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THE 12 MICRON BAND OF ETHANE: A SPECTRAL CATALOG FROM 765 CM⁻¹ TO 900 CM⁻¹

A. K. Atakan, W. E. Blass, J. W. Brault, S. J. Daunt, G. W. Halsey, D. E. Jennings, D. C. Reuter and J. Susskind

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Goddard Space Flight Center Greenbelt, Maryland 20771

Abstract

This document is concerned with the high resolution laboratory absorption spectrum of the $12\mu m$ band of ethane gas. The data were obtained using the McMath Solar Telescope 1-metre Fourier Transform interferometer at Kitt Peak National Observatory and tunable diode laser spectrometers at the University of Tennessee and NASA/Goddard Space Flight Center. Over 2000 individual vibration-rotation transitions were analyzed taking into account many higher order effects including torsional splitting. Line positions were reproduced to better than 0.001 cm $^{-1}$. Both ground and upper state molecular constants were determined in the analysis.

Part I of this document contains a discussion of the experimental details, the analysis procedures and the results. A list of ethane transitions occurring near $^{14}\text{CO}_2$ laser lines needed for heterodyne searches for C_2H_6 in extraterrestrial sources is also included.

Part II contains a spectral catalogue of the ethane v_9 fundamental from 765 cm⁻¹ to 900 cm⁻¹. The contents include: (a) a high dispersion (1 cm⁻¹/12 in.) plot of both the Kitt Peak interferometric data and a simulated spectrum with Doppler-limited resolution using the model of Part I; (b) a table of over 8500 calculated transitions listing quantum number assignments, frequencies and intensities.

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PART I

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The $12\mu m$ Band of Ethane: High Resolution Laboratory Analysis with Candidate Lines for Infrared Heterodyne Searches*

S.J. Daunt¹
Concordia University/University of Tennessee

A.K. Atakan, W.E. Blass and G.W. Halsey University of Tennessee

D.E. Jennings², D.C. Reuter³ and J. Susskind NASA/Goddard Space Flight Center

and

J.W. Brault Kitt Peak National Observatory

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¹Natural Sciences and Engineering Research Council of Canada University Research Fellow 1980-85.

²Guest observer at Kitt Peak National Observatory, operated by the Association of Universities for Research in Astronomy, Inc., under contract with the National Science Foundation.

³National Research Council/Resident Research Associate (1981-82).

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ABSTRACT

Several groups have reported emission from the v_q band of ethane at $12\mu m$ in the spectra of Jupiter, Saturn, Neptune and Titan. This paper reports the results of a laboratory study of v_9 using both high resolution Fourier Transform and diode laser absorption spectroscopy. Approximately 2000 transitions in this band have been subjected to an analysis that includes the normal rovibrational terms as well as the higher-order effects of ℓ -doubling, l-resonance, internal rotation and a Coriolis resonance with the 304 state. A model for this band capable of reproducing observed $\,\nu_{9}$ features to better than 0.001 cm⁻¹ is presented. High precision values of the primary ground state constants have also been determined: $B_0 = 0.6630279(24) \text{ cm}^{-1}$, $D_0^{J} =$ $1.0324(23) \times 10^{-6} \text{ cm}^{-1} \text{ and } D_0^{JK} = 2.651(88) \times 10^{-6} \text{ cm}^{-1}$. A list of v_9 transitions occuring near $^{14}\mathrm{CO}_2$ laser lines that are good candidates for laser heterodyne searches has been compiled. An atlas is available from the authors that includes: (1) plots of the observed and simulated ν_9 spectra between 750 and $900~\text{cm}^{-1}$ and (2) a list of over 8500 calculated transitions and intensities.

I. INTRODUCTION

The ν_9 fundamental band of ethane occurs in the $12\mu m$ wavelength region and has recently received a great deal of attention in molecular astrophysics. It is the strongest band of ethane in a terrestrial window and therefore has always provided the best opportunity for identification and characterization of ethane in planetary atmospheres and molecular clouds.

Spectra recorded from the outer solar system bodies of Jupiter, Saturn, Neptune and Titan show 12µm emission from ethane (Ridgway 1974; Gillett and Forrest, 1974; Combes et al., 1974; Tokunaga, Knacke and Owen, 1976; Encrenaz, Combes and Zéau, 1978; Tokunaga et al., 1979; Tokunaga, Knacke and Orton, 1975; Gillett, 1975; Gillett and Rieke, 1977; Macy and Sinton, 1977; Gillett, Forrest and Merrill, 1973; Hanel et al., 1981). The observed v_9 emission has been used to determine ethane abundances and to map planetary ethane (Gillett and Orton, 1975; Tokunaga et al., 1978; Hanel et al., 1981). All of these studies provide information needed for the construction of planetary atmospheric models (Danielseon, et al, 1973; Hunten, 1974; Strobel, 1974; Caldwell, 1977). In low resolution spectra the emission is seen as a single $\sim 70~{\rm cm}^{-1}$ wide feature while its appearance in medium resolution spectra is as a series of unresolved Q-branches separated by ~2.7 cm⁻¹. Very recently, however, high resolution laser heterodyne spectroscopy has detected individual vibration-rotation lines of v_q in emission from Jupiter's stratosphere (Kostiuk et al., 1981 and 1983). This laser heterodyne astronomy program was a major reason for undertaking the present laboratory study.

Although C_2H_6 has not been detected in molecular sources outside the solar system, the $12\mu m$ band is again the best candidate for a search. Ethane

cannot be detected in the radio region because it has no permanent dipole moment 1 . However, infrared laser heterodyne spectroscopy has recently provided the $10\mu m$ detection of the similarly nonpolar ethylene molecule (C_2H_4) in the circumstellar cloud of IRC + 10216 (Betz, 1981). Two other non-polar hydrocarbons, methane and acetylene, have already been identified in the infrared spectrum of this source (Hall and Ridgway, 1978; Ridgway et al., 1976). The overlap of the $^{14}C^{16}O_2$ laser with the R-side of v_9 means that laser heterodyne searches can be made for ethane in IRC + 10216 and other infrared molecular sources.

The application of infrared laser heterodyne spectroscopy to observations of high-altitude ethane on Jupiter and the possibility that this technique will be used to search for ethane line emission in other planets and molecular clouds makes a new laboratory analysis of v_9 important at this time. In the previous study (Daunt et al., 1981) the rotational structure in the Q-branches was not resolved. In particular, line-splittings due to torsional effects and frequency shifts and intensity anomalies due to higher order Coriolis effects were not observed. Absolute line frequencies measured in that work were only accurate to about 0.01 cm⁻¹. The narrow bandwidth, high resolution and frequency precision of laser heterodyne techniques requires individual line positions to be known to better than 0.001 cm⁻¹.

 $^{^1\}text{Pure}$ rotational transitions are made weakly allowed through interactions between the torsional mode and dipole allowed vibrations as discussed by Rosenberg and Susskind (1979). They have predicted frequencies and intensities for ΔK = 0, ΔJ = ± 1 transitions which would occur in the millimeter wave region. There has been no experimental detections of these lines reported to date.

A new study of the ν_9 band of ethane was therefore undertaken using both high resolution Fourier transform and tunable diode laser spectra. The combination of these two types of spectroscopy yielded complete coverage of the band with high absolute frequency precision and complete resolution (to the Doppler linewidth and beyond) in dense spectral regions. The results of a full vibration-rotation analysis including higher order effects is reported here. This analysis allowed accurate line frequencies and intensities to be calculated. An independent study of ν_9 by Henry <u>et al</u>. (1983) has yielded results similar to those reported in the present paper.

II. EXPERIMENTAL DETAILS

The ethane spectra were recorded with the one-meter Fourier transform spectrometer (FTS) located at the McMath solar telescope at Kitt Peak National Observatory. This instrument is a dual, moving cat's-eye interferometer capable of 0.005 cm⁻¹ unapodized resolution when operated in the asymmetric interferogram mode. The beamsplitters used for this work were made of KBr and the detector was a liquid helium cooled Arsenic-doped Silicon photoconductor. A 450 Watt glower was used as a continuum source. The reference signal for this instrument is provided by a stabilized HeNe laser. Thirty-two interferograms were averaged during a period of 103 minutes to produce a S/N ratio of approximately 400/1.

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The ethane was a research-grade sample (99.99%) obtained from Matheson Gas Products and used without purification. The gas was contained in a 30 cm straight-path cell with BaF $_2$ windows. The sample pressure was 4.01 Torr, as measured with a capacitance manometer. The temperature of the sample cell was 22°C. Absolute frequency calibration was established by recording the C_2H_6 spectrum simultaneously with a spectrum of CO_2 . Carbon dioxide at a pressure of 0.5 Torr was contained in a White-type multiple traversal cell set for a path length of 193m. The source radiation beam was passed simultaneously through both cells before entering the spectrometer. The 10μ m $^{12}C^{16}O_2$ laser frequencies of Petersen et al. (1974) were used as standards, and errors introduced by extrapolating the calibration to 12μ m are estimated to be less than 2×10^{-4} cm $^{-1}$.

The line splittings due to torsional and torsional-Coriolis effects were studied using tunable diode laser spectra. The experimental details of that

portion of the work have been described in a separate paper (Susskind <u>et al.</u>, 1982). The diode laser results were used in the present work to measure absolute line intensities and to model the splittings and line shifts which were not sufficiently resolved in the FTS spectra.

III. ANALYSIS

(a) Assignment of Spectra

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The FTS spectra were assigned by taking the molecular parameters determined for v_9 from the previous study of Daunt et al. (1981) and generating a simulated spectrum with programs described in that work. A list of quantum number assignments, wavenumbers and relative intensities was generated along with a computer plot of the spectrum. This calculated spectrum with assignments was then used to identify the features in the observed spectrum. The earlier work had clearly resolved only the Pp and RR lines of v_9 . The new FTS data resolved the ^{R}Q and ^{P}Q branches as well as the ^{R}P and ^{P}R lines in addition to the ^{P}P and ^{R}R structure. Despite the limited selection of lines in the previous study the parameters determined there gave an excellent prediction of the FTS spectrum. The upper trace of Figure 1 shows the $^{R}Q_{\Delta}$ region of v_{9} as recorded by the Kitt Peak FTS instrument. The lower trace is a simulation at $0.005~{\rm cm}^{-1}$ resolution using the constants from Daunt et al. (1981). The structure of this Q-branch had not been resolved in the earlier work (see Fig. 2 of Daunt et al., 1981), yet the prediction was excellent, enabling assignments to be made in a straightforward manner. Any questionable assignments were checked by the traditional method of combination differences.

(b) <u>Theoretical Model</u>

The vibration-rotation Hamiltonian for the ethane molecule can be written to first order as

$$H = H_{VIB} + H_{ROT} + H_{VR}$$
 (1)

where H_{VIB} and H_{ROT} are the vibration and rotation Hamiltonians for a symmetric rotor (Herzberg, 1945; Allen and Cross, 1963). The term H_{VR} represents vibration-rotation coupling contributions including the usual Coriolis interactions described by Mills (1972). The rovibrational Hamiltonian, Eq. (1), used in this work contained all terms through h_3^{\dagger} plus the β terms of h_4^{\dagger} as described by Blass and Nielsen (1974) and Daunt <u>et al</u>. (1981):

$$\langle J, K, \ell_{9} | H | J, K, \ell_{9} \rangle = \nu_{0} + (A_{0} - B_{0}) K^{2} - 2A_{e} \zeta_{9}^{Z} K \ell_{9}$$

$$+ B_{0} J (J+1) - (\alpha_{9}^{A} - \alpha_{9}^{B}) K^{2} - \alpha_{9}^{B} J (J+1) - D_{0}^{J} J^{2} (J+1)^{2}$$

$$- D_{0}^{JK} K^{2} J (J+1) - D_{0}^{K} K^{4} + n_{9}^{J} J (J+1) K \ell_{9}$$

$$+ n_{9}^{K} K^{3} \ell_{9} + \beta^{J} J^{2} (J+1)^{2} + \beta^{JK} K^{2} J (J+1) + \beta^{K} K^{4}$$

$$(2)$$

The previous study of Daunt et al. (1981) showed that ℓ -doubling and ℓ -resonance interactions were necessary for understanding the observed features of ν_9 . The necessary matrix elements have been given by Amat, Nielsen and Tarrago (1971) as:

$$\langle J, K+1, \ell_9 = +1 \mid H \mid J, K-1, \ell_9 = -1 \rangle =$$

$$-\frac{1}{2} \left[q_9^{(+)} + \delta q_9^J J(J+1) + \delta q_9^K (2K)^2 \right]$$

$$\left[\{ J(J+1) - K(K+1) \} \{ J(J+1) - K(K-1) \} \right]^{\frac{1}{2}}$$

$$(3)$$

where the following definitions relative to Amat, Nielsen and Tarrago (1971, Table XXXVI) have been used in the present work:

$$q_{t}^{(+)} = -4F_{t}^{t}$$

$$\delta q_{t}^{J} = -4F_{22+}^{t,J+}$$

$$\delta q_{t}^{K} = -8F_{22+}^{t,K+}$$
(4)

The sign convention used for the ℓ -doubling constant $q_t^{(+)}$ is the generally accepted one of Cartwright and Mills (1970).

The earlier lower resolution study (Daunt \underline{et} al., 1981) required only those terms described by Eqs. (2)-(4). Torsional splittings and higher order effects were not needed to account for the observed data and were therefore not included in the model.

The characteristics of torsional spectral features in ethane were first referred to by Wilson (1938) and later were discussed in more detail by Susskind (1974) and Hougen (1980). Observed ethane torsional splittings were first reported by Flicker et al. (1977) in the ν_9 band but went unanalyzed at that time. Pine and Lafferty (1982) observed torsional splittings in the 3μ m bands but the strong interactions among the many vibration-rotation states prevented a quantitative upper state analysis. Patterson et al. (1981) correctly interpreted the observed intensity anomalies in the diode laser spectrum of the RQ_0 branch of ν_9 as due to the effects of torsional splittings. Diode laser spectra recorded by the present authors across a wide range of the ν_9 band and described in more detail elsewhere (Susskind et al., 1982) clearly showed torsional splittings plus the effects of a Coriolis interaction with the $3\nu_4$ torsional state.

These newest observations indicated that the high resolution FTS data needed to be analyzed with a model that included torsional effects. This changed the Hamiltonian of Eq. (1) to

$$H = H_{VIB} + H_{ROT} + H_{VR} + H_{TOR} + H_{TV}$$
 (5)

where the details of the new terms H_{TOR} and H_{TV} are described elsewhere (Susskind <u>et al.</u>, 1982; Susskind, 1974; Hougen, 1980). Only the most important effects of these terms on the appearance of the spectra will be discussed here.

The torsional term H_{TOR} causes each $|J,K,\ell\rangle$ vibration-rotation state to be split into four torsional substates denoted by the torsional symmetry number $\sigma=0$, 1, 2 or 3. As shown in Fig. 2 the allowed levels in the ground vibrational state have the following value relationship: even σ for K even and odd σ for K odd. In the $v_9=1$ state the reverse is true. The overall selection rules for v_9 transitions are $\Delta K=\pm 1$, $\Delta \ell=\pm 1$, $\Delta J=0$, ± 1 and $\Delta \sigma=0$. The H_{TV} term allows each vibrational state to have a different value for the barrier to internal rotation. To the extent that this barrier changes, the spacings of the σ torsional levels will differ for each vibrational state.

Fig. 2 also shows the splitting (Δ_9) between the allowed torsional levels in ν_9 to be less than that in the ground state (Δ_G) . If such a difference between the torsional level splittings in lower and upper states exists, then each normally allowed transition, denoted by the spectroscopic symbol $^{\Delta K}[\Delta J]_K(J)$, will be split into a torsional doublet. The first order torsional model given by Susskind (1974) predicts that all ν_9 doublets should have essentially the same splitting of $S_{0,9} \equiv |\Delta_9 - \Delta_G|$. One torsional component is shifted ΔS (= $S_{0,9}$) from the unsplit position while the other component is shifted 2 ΔS in the opposite direction. The sense of the shifts depend on the appropria+ round state K values as given in Table I. The

diode laser spectra (Susskind et al. 1982) showed that the torsional doublets at low J and K values were indeed split by a constant value of $^{-2}.2 \times 10^{-3}$ cm $^{-1}$ but that lines with higher J and K values sometimes had splittings of more than 0.05 cm^{-1} . This observation could not be accounted for using only the terms H_{TOR} and H_{TV} . An accidental Coriolis resonance with the nearby torsionally excited state $3\nu_4$ was shown to account for the observed J,K-dependent splitting in excess of the intrinsic torsional splitting, $S_{0,9}$. This had the effect of perturbing each component of a doublet by a different amount, thereby altering the apparent splitting. A final model Hamiltonian including all the important effects is then given by

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$$H = H_{VIB} + H_{ROT} + H_{VR} + H_{TOR} + H_{TV} + H_{COR}$$
 (6)

where H_{COR} is described in detail in Susskind <u>et al</u>. (1982). The energy levels involved in this resonance are schematically shown in Fig. 3. The $3\nu_4$ state has four torsional σ substates with the vibrational origin energies shown in the figure. The energy separations from the origin of the ν_9 state are given by $\Delta_{\sigma} = \nu_9 - (3\nu_4)_{\sigma} = \Delta_0$, Δ_1 , Δ_2 and Δ_3 . It should be noted from this figure that the $|\nu_9, J, K-1, -1\rangle$ levels (-£ states, see Herzberg, 1945) are closer to the $3\nu_4$ levels than the $|\nu_9, J, K+1, +1\rangle$ levels (+£ states). Since the selection rule for the Coriolis interaction is $K_{\nu_9} - K_3 \nu_4 = \pounds_{\nu_9}$ the perturbations are expected to be greater for the -£ levels than for the +£ levels. The observed diode laser spectra of the ${}^PQ_K(J)$ branch ($\Delta K = \Delta \pounds = -1, \Delta J = 0$) lines showed splittings that were much more perturbed than those for the ${}^RQ_K(J)$ branches ($\Delta K = \Delta \pounds = +1, \Delta J = 0$) in agreement with this model. A diagram of how the individual vibration-rotation-torsion states are coupled through the Coriolis resonance is given in Fig. 4 for the case of K'' = 6. The magnitude of

the perturbation to any $|v_9,J,K+\ell,\ell,\sigma\rangle$ level through H_{COR} is given by second-order perturbation theory as

$$\Delta E = \frac{C^{2}[J(J+1)-K(K+\ell)]}{\Delta_{\sigma} + E^{\nu_{9}}_{J,K+\ell,\ell} - E^{3\nu_{4}}_{J,K}}$$
(7)

where C is a coupling constant described in Susskind <u>et al</u>. (1982) and found to have the value $0.032~\rm cm^{-1}$. The perturbation is different for each σ torsional state. Therefore the total splitting for a transition to an upper state $|J,K+\ell,\ell\rangle$ is given by

$$S_{J,K+\ell,\ell} = S_{0,9} + C^2 [J(J+1) - K(K+\ell)]F(J,K,\ell)$$
 (8)

where

$$F(J,K,\ell) = \frac{1}{\Delta_{\sigma} + E_{J,K+\ell,\ell}^{\vee 9} - E_{J,K}^{3\nu_{+}}} - \frac{1}{\Delta_{\sigma+2} + E_{J,K+\ell,\ell}^{\vee 9} - E_{J,K}^{3\nu_{+}}}. (9)$$

The rotational contribution to each denominator in Eq. (9) is given to first order by

$$E_{J,K+\ell,\ell}^{\vee 9} - E_{J,K}^{3\vee_{4}} = 2[A(1-\zeta_{9}^{2})-B]K\ell + [A(1-2\zeta_{9}^{2})-B]$$

$$= 2.617K\Delta K + 0.619 \text{ cm}^{-1}$$
(10)

The primary source of any observed splitting is given by the first term on the r.h.s. of Eq. (8) i.e., the intrinsic splitting: $S_{0,9}$. The second factor affecting the observed splittings comes from the difference in the Coriolis perturbations to each component as given by the second term on the r.h.s. of Eq. (8). The intrinsic splitting $S_{0,9}$ ($\approx 2.2 \times 10^{-3} \text{ cm}^{-1}$) dominates far from

resonance, i.e., for lines having large K" Δ K values or those near the subband origins (JzK). For lines with either larger (J-K) or more negative K" Δ K values the Coriolis effects become progressively greater. This results in frequency shifts which enlarge the torsional component splittings to values sometimes exceeding 0.050 cm $^{-1}$. The Coriolis perturbation is positive and always greater for the σ (=0 for K even and =1 for K odd) levels than for the σ + 2 levels provided the interacting ν_9 and $3\nu_4$ states do not cross. This is in the same sense as the first order torsional splitting, $S_{0,9}$, assuming a barrier in ν_9 greater than that in the ground state. Since all the lines treated in this paper were not near the level crossing (K" Δ K=-17) the Coriolis perturbation always increased the splittings.

(c) Correction of the Data Set

The original FTS data set consisted of over 2000 assigned lines having quantum number values of $-13 \le K'' \triangle K \le +18$ and $J'' \le 40$. These lines covered the spectral region between 765 cm⁻¹ and 900 cm⁻¹ and at first inspection did not show torsional splittings. This is understandable since in the previous section it was seen that the first order splitting, $S_{0,9}$, was 0.0022 cm⁻¹ and the unapodized resolution of the FTS data was 0.005 cm⁻¹. Lines with high J and K quantum numbers have observable splittings greater than the resolution only when the Coriolis contribution of Eq. (8) is sufficiently large. These enhanced splittings are visible on careful inspection of the data. However, the resolution and S/N ratio of the data set used for the present study was such that most of the torsional doublets were either unresolved or the weaker component was not reliably measured.

The very high frequency precision of the FTS data meant that some correction for the combined instrumental and torsional effects (splittings and shifts) had to be carried out before a complete analysis would yield reliable molecular constants free of systematic model errors. All the terms of Eq. (6) can be treated simultaneously using a global model Hamiltonian but this was deemed unnecessary since the torisonal terms are essentially independent of the rest of the terms in Eq. (6). Therefore an alternative method to full diagonalization was used. The effects of torsional splitting and the Coriolis resonance with 304 were calculated using Table I and Eq. (7), respectively. These calculations were then used, together with the instrumental response, to deperturb the observed data yielding "corrected" frequencies which could be fitted with Eqs. (2) and (3). The clearest way to describe this procedure is to refer the reader to Fig. 5. The torsional splitting from the unperturbed and unsplit position (ν_0) was calculated according to Table I using the results from Susskind et al. (1982) (Fig. 5a). The effects of the Coriolis resonance with $3v_4$ on each torsional component was then calculated using Eq. (7) and added to the first order torsional result (Fig. 5b). This was followed by giving each component transition a Doppler width profile (Δv_D = $1.8 \times 10^{-3} \text{ cm}^{-1}$ FWHM) in the proper intensity ratio (Table I) and adding the result in the case of overlap (Fig. 5c). The next step was a convolution with the appropriate instrument profile. This was followed by a peak finding routine that located maxima in the final profile and calculated the shift(s) from the origin ν_0 (Fig. 5d). These shifts were then subtracted from the observed transition frequencies to yield a corrected data set. The modified data set then corresponds to the spectrum of ν_{Q} which would be observed if torsion and the $3v_4$ resonance did not exist. This corrected data set does not represent any physically observable spectrum but it allows the fitting of the band to a standard model that simplifies both the analysis and subsequent

simulation of the spectrum for astrophysical modeling. Simulated spectra for comparison with observed data are easily obtained by adding the calculated torsional and Coriolis effects to the unsplit line positions to predict the final spectrum.

(d) Fitting Procedure

The data analysis procedure followed the iterative scheme listed below:

- (1) each 2x2 Hamiltonian (Cartwright and Mills, 1970) was evaluated for a molecular parameter set of 1 parameters $[\gamma_1, \gamma_2, \ldots, \gamma_1]_n = \tilde{\gamma}_n$ (n = 0 for the initial set) to obtain the upper state energies;
- (2) the residuals (obs.-calc.) were computed as

$$\Delta_{O-C} (J+\Delta J, K+\Delta K, \ell+\Delta \ell) = [\nu_{OBS}(J+\Delta J, K+\Delta K, \ell+\Delta \ell)]$$

+
$$E_{gs}(J,K,\ell=0)$$
] - $E_{calc}^{n}(J+\Delta J,K+\Delta K,\ell+\Delta \ell)$ (11)

where E_{qs} is the ground state energy and E_{calc}^{n} came from step (1);

- (3) the derivatives $\delta E/\delta \gamma_m$, where γ_m is a particular molecular parameter such as α_9^B , were calculated using the Hellman-Feynman theorem (Rowe and Wilson, 1975);
- (4) the residuals, Δ_{o-c} , were fitted to a Taylor series expansion of the energy retaining only the terms linear in $\delta \gamma$, i.e.

$$\Delta_{O-C} = \sum_{m} \frac{\delta E}{\delta \gamma_{m}} \Big|_{\widetilde{\gamma}_{n}} \delta \gamma_{m} \qquad ; \qquad (12)$$

(5) the molecular parameter set $\widetilde{\gamma}_n$ was corrected according to

$$(\gamma_{m})_{n+1} = (\gamma_{m})_{n} + \delta \gamma_{m}$$
 (13)

The series of steps from 1 through 5 continued until the stepwise regression (Efrymonson, 1960) produced no significant reduction in the sum square error of the remaining $\Delta_{\rm O-C}$ of step 2. At that point, the data, which had been originally weighted uniformly (wt.=1), was reweighted according to the method of bi-weights (Beaton and Tukey, 1974) as in the previous ν_9 study (Daunt et al., 1981). The procedure was terminated using a modified χ^2 test (Lin et al., 1980).

(e) Results

Several parameter sets, corresponding to model Hamiltonians limited to lower order terms than those presented in Eqs. (2) and (3), were also tried in the analysis procedure described above. In all, four parameter sets were determined involving inclusion and exclusion of $\{\beta^J, \beta^K, \beta^K\}$ and $\{\delta q_g, \delta q_g^K\}$ in various combinations. Each parameter set was used to synthesize plots of selected 2 cm $^{-1}$ segments of the spectrum on a 1 cm $^{-1}$ /meter scale. These were then compared with the observed spectrum plotted on the same scale. Based on these visual comparisons, the best parameter set was judged to be the full set described by Eqs. (2) and (3) and listed in Table II. The analysis of 1609 corrected transitions yielded a standard deviation of 0.7 x $10^{-3} \ \mathrm{cm}^{-1}$. The overall agreement between the ν_9 molecular constants of the present study and those reported by Daunt et al. (1981) and Henry et al. (1983) is very good. The only notable discrepancy occurs in the band origin ν_{0} which appears to be $\sim 0.001~{
m cm}^{-1}$ too low in Henry et al. (1983). The fact that their interferometer has to be operated in air at present is probably responsible for this small but significant wavelength offset.

A ground state constant determination was also carried out as in Lin et al. (1980) using 893 ground state combination differences. Results of this analysis and comparison with previous work are shown in Table III. The excellent agreement with the analyses of laser difference frequency data by Pine and Lafferty (1982) and recent FTS data by Henry et al. (1983) indicates that the ground state constants for ethane are now very precisely determined.

The quality of the analysis is best demonstrated by examination of Figures 6-9 which show the ${}^RQ_K(J)$, K=1, 2, 3 and 4 branch regions of v_9 . Panel A of each figure shows the FTS data and panel B a corresponding diode laser scan of the same region.

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The Doppler limited diode laser data was computer deconvoluted using the techniques described by Blass and Halsey (1981) in order to clearly resolve the torsional splittings at low J and K values. These results are shown in panel C of each figure. The intrinsic torsional splitting of $S_{0.9} = 2.2 \text{ x}$ $10^{-3}~{\rm cm}^{-1}$ is now readily visible. The parameters of Tables II and III for the deperturbed vg plus the torsional/Coriolis parameters of Susskind et al. (1982) were then used to simulate the deconvoluted spectra. The results for a calculated line width of $0.001~\text{cm}^{-1}$ are shown in panel D of each figure. The agreement is most pleasing in each case. Features not reproduced are attributed to the hot band $v_9 + v_4 - v_4$ which has not been included in the simulation. It is evident from Figs. 6-9 that the present model can reproduce essentially all the observed vo features in the present data with good frequency accuracy (better than 0.001 cm⁻¹). The relative intensities of the torsional doublets in Figs. 6-9 appear to deviate from the calculated because the scans available for deconvolution were not recorded under conditions necessary for accurate absolute intensity measurements. Additional diode

laser spectra recorded for the purpose of intensity measurements gave excellent agreement with the theoretical predictions of Table I.

IV. DISCUSSION

One of the most important features of the ν_9 band is the $^RQ_0(J)$ branch. This band head has been used to evaluate the ethane abundance in the Jovian atmosphere (Tokunaga, Knacke and Owen, 1976; Tokunaga et al., 1979) because it stood out clearly from the remainder of the band. It was noted, however, by Tokunaga and Varanasi (1976) that it was impossible to simultaneously fit RQ_0 and the rest of the ν_9 band to a standard model. These workers obtained effective molecular parameters for RQ_0 and made laboratory intensity measurements which were then used to extract mixing ratios and abundances from the astrophysical spectra.

The anomalous behavior of RQ_0 is due to 2-doubling effects (Daunt et al., 1981) induced by the Coriolis interaction of v_9 with v_4 , v_5 and v_6 (Susskind et al., 1982). This explains the difference between RQ_0 and the other $^P, ^RQ_K$ branches reported by Tokunaga and co-workers (1976, 1979). The detailed line pattern of RQ_0 was partially resolved in the diode laser spectra of Flicker et al. (1977) and interpreted correctly later by Patterson et al. (1981). Only when taking the torsional effects into account could the unexpected intensity pattern of the lines be understood. The effect of the Coriolis interaction on the component splitting is a factor of two larger for RQ_0 than for most other branches (the exceptions being RR_0 and RP_0 where there is no torsional—Coriolis interaction). The combination of this enlarged torsional splitting and the narrowing of J-line spacings caused by the 2-doubling are responsible for the anomalous appearance of RQ_0 . This feature is the final test for the present model. Figure 10A shows the FTS spectrum where the line spacings are so close that very little is resolved. A Doppler limited diode laser scan of

 $^R\mathbb{Q}_0$ is shown in Fig. 10B where more structure is visible but still not enough for a detailed line-by-line analysis. If the spectrum in Fig. 10B is deconvoluted, then complete resolution of all the torsional features is obtained as seen in Fig. 10C. The peculiar pattern is due to the lower frequency torsional component of each J falling into close coincidence with the high frequency component of the next higher J. The complete model for ν_9 predicts the pattern plotted in Fig. 10D and is in good agreement with the deconvoluted laser data. The extra lines appearing in Fig. 10C are assumed to be transitions of the hot band ν_9 + ν_4 - ν_4 .

An accurate model of the line positions and relative intensities of $^{R}Q_{0}$ is now available. The uncertainty of earlier astrophysical studies caused by a lack of understanding of the structure of this feature has been removed.

There have been several attempts to obtain absolute intensity values for the entire v_9 band (Varanasi, Cess and Bangaru, 1974) and for the RQ_0 branch alone (Tokunaga and co-workers, 1976; 1979). A combination of our calculated relative line strengths and diode laser absolute line strength determinations has allowed us to derive a value of 0.53 (\pm 0.05) cm $^{-2}$ atm $^{-1}$ at 296°K for the RQ_0 branch. The integrated strength from the FTS data for the RQ_0 region gave S = 0.56 (\pm 0.03)cm $^{-2}$ atm $^{-1}$. These values are much lower than the S = 0.74 (\pm 0.09) cm $^{-2}$ atm $^{-1}$ obtained by Tokunaga and Varanasi (1976) from unresolved medium resolution data. The value of 0.70 cm $^{-2}$ atm $^{-1}$ derived from diode laser data reported in Henry et al. (1983) is more problematic. They used a rigid rotor model and measured only RQ type transitions with low K and high J quantum numbers. These are the lines, however, whose intensities are the most perturbed by the &(2,2) resonance. The extent of this perturbation is quite pronounced as shown in Table IV and Figure 11. The RQ_1 (25) and RQ_1 (30) lines are 22% and

31% stronger than normal (rigid rotor model) and will appear to be 44% and 62% stronger than the respective $^{P}Q_{1}(25)$ and $^{P}Q_{1}(30)$ lines (see Fig. 11).

The K and J dependence of this effect accounts for the problems reported in Henry et al. (1983) in trying to derive a consistent transition dipole moment as K and J changed. These types of intensity effects have been described in more detail in Mills (1969), Cartwright and Mills (1970) and Masri and Blass (1971). In short, the overall intensity effect of the resonance (for the case of v_9 of C_2H_6) is to enhance RQ , PP and PR type transitions while depleting the PQ , RR and RP type transitions. Unless one samples all sets of transitions for wide values of K and J the derivation of a band strength will be in error. It would be possible to derive strengths from a few selected lines but only if the resonances have been rigorously taken into account. The particular case of v_9 of C_2H_6 strongly illustrates the inadequacy of using rigid rotor models for the study of perturbed bands.

In comparing the two diode laser line intensities studies where measurements overlap the present study obtained experimental strength values about 40% lower than those reported in Henry et al. (1983). This disagreement is independent of the model problem discussed above. Since large discrepancies still exist, further work on the absolute intensity problem is required before $^{R}Q_{0}$ or any other features of v_{9} can be used as precise probes for ethane abundances. It should also be remembered that a large proportion of the observed intensity in the $12\mu m$ region is attributable to the as yet unanalyzed hot bands (v_{9} + nv_{4} - nv_{4}), the $3v_{4}$ band, and isotopic bands. Work on these problems is presently underway.

Recently Bjoraker <u>et al.</u> reported a detection of ethane on Saturn in the $3\mu m$ region through a band strikingly similar in appearance to ν_9 (Q-branch

spacings ~2.7 cm $^{-1}$ apart). There is also the possibility of using the v_9 + v_{12} band (Lin, Blass and Gailar, 1980) as a probe for ethane as suggested by Treffers et al. (1978). However, v_9 would still appear to be the strongest feature of C_2H_6 in the usual atmospheric windows and therefore the best candidate for extraterrestrial searches.

The suitability of the vo band for laser heterodyne searches in molecular sources was a primary motivation of the present study. The R side of the v_9 band overlaps the I-band P-branch of the $^{14}\mathrm{CO}_2$ laser lines and provides the best opportunities for heterodyne studies. Assuming a Doppler shift for a source of ±3GHz (as for Jupiter) and a filter bank width of ±3GHz, candidate coincidences within $\pm 6 \text{GHz}$ ($\pm .2 \text{ cm}^{-1}$) between the stronger transitions of v_9 and the $^{14}\text{CO}_2$ laser lines are listed in Table V. The quality of the reported offsets have been checked recently by laboratory heterodyne measurements by Weaver et al. (1983). Comparisons with the predictions in Table V for the eight lines studied show an average difference of only 6MHz. Several of these predicted coincident ethane transitions have recently been observed in the Jovian stratosphere by Kostiuk et al. (1983). The relative intensities listed in Table IV can be converted to absolute intensities by applying the multiplicative conversion factor 7.1(0.7) \times 10⁻³ cm⁻² atm⁻¹ This factor was derived from diode laser intensity measurements of v_9 transitions and corresponds to a sample temperature of 296°K.

The lines listed in Table V have been taken from an atlas of calculated v_9 transitions compiled from the present study and covering the region from 750 to 900 cm⁻¹. This atlas consists of two sections: Part I - plots of the observed FTS spectrum and a simulated spectrum calculated at Doppler width for 300°K (1.8 x 10^{-4} cm⁻¹); and Part II - a list of over 8500 calculated

transition frequencies, quantum number assignments and relative intensities. A page from Part I of this atlas for the 837.5 to 840.0 cm⁻¹ region is shown in Fig. 12. The corresponding page list of Part II for the plots in Fig. 12 is given in Table VI. The intensity conversion factor discussed above applies to the entire ν_9 atlas. Copies of this atlas may be obtained by writing to the authors (S.J.D., D.E.J. or W.E.B.).

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ADDRESSES OF THE AUTHORS

- S.J. Daunt: Department of Chemistry, H-1139, Concordia University, 1455 de Maisonneuve Boulevard West, Montréal, Québec, Canada H3G 1M8; and Department of Physics and Astronomy, The University of Tennessee, Knoxville, TN 37996-1200.
- A.K. Atakan, W.E. Blass and G.W. Halsey: Molecular Spectroscopy Laboratory, Department of Physics and Astronomy, The University of Tennessee, Knoxville, TN 37996-1200.
- D.E. Jennings: Infrared and Radio Astronomy Branch, Laboratory for Extraterrestrial Physics, Code 693, NASA/Goddard Space Flight Center, Greenbelt, MD 20771
- D.C. Reuter and J. Susskind: Goddard Laboratory for Atmospheric Sciences, Code 911, NASA/Goddard Space Flight Center, Greenbelt, MD 20771
- J.W. Brault: Kitt Peak National Observatory, Tucson, AZ 85726

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FIGURE CAPTIONS

- Figure 1. Upper panel: part of the FTS spectrum of the v_9 band of C_2H_6 near R_0 4 recorded with $\Delta v = 0.005$ cm $^{-1}$. Lower panel: simulated spectrum of the same region using the parameters from Daunt <u>et al.</u> (1981). No torsional effects were included in this calculation.
- Figure 2. Selection rules, first order torsional splittings, predicted splitting patterns and intensities for an internal rotation barrier in ν_9 greater than that in the ground vibrational state. The allowed and forbidden levels are shown by horizontal solid and dashed lines, respectively.
- Figure 3. Energy origins of the $3\nu_4$ state (including torsional effects) relative to ν_9 . Note that the (-1) states of ν_9 are closer to the $3\nu_4$ levels than the (+1) states. Since transitions with $\Delta K = -1$ (PP , PQ , PR) terminate in the (-1) levels, they will be the most perturbed by $3\nu_4$.
- Figure 4. Energy levels involved in transitions from the ground state (K" = 6, J") to ν_9 . Torsional level splittings are shown for the lower (+2S₀, +S₀, -S₀, -2S₀ where S₀ = $\Delta_G/3$) and upper states (+2S₁, +S₁, -S₁, -2S₁ where S₁ = $\Delta_9/3$). The allowed torsional levels are shown by solid lines and the forbidden levels by dashed lines. The $3\nu_4$ levels perturb the ν_9 levels through the Coriolis interaction matrix shown in the upper left corner (the interacting levels are connected by the lines ••••). Allowed ν_9 transitions are shown by the slanting vertical arrows.
- Figure 5. Method used for obtaining corrective shifts for the FTS data.

- (a) Calculate the first order torsional splittings for each ν_9 transition (unperturbed frequency = ν_0). The pattern is given by Table I where $S_{0,9}=|3\Delta S|=|3(S_1-S_0)|=|\Delta_9-\Delta_G|=2.282\times 10^{-3}$ cm⁻¹. (b) The $3\nu_4$ Coriolis resonance contributions are calculated with Eq. (7) and these additional shifts are added to each torsional component.
- (c) Each line is given a Doppler line shape ($\Delta v_D = 1.8 \times 10^{-3} \text{ cm}^{-1}$ FWHM) and intensities are summed. (d) The results from part (c) are convoluted with an instrument function to yield a final contour. This profile is then searched for maxima and shifts relative to v_0 are calculated. These shift values are then merged with the observed measurements to create an "unsplit and unshifted" set of frequencies that can be fitted with the standard model of Eqs. (2)-(3).
- Figure 6. The $^{R}Q_{1}$ region of v_{9} . (A) FTS spectrum; (B) diode laser scan; (C) deconvoluted diode laser data; (D) simulated spectra including torsion and Coriolis effects (full model).
- Figure 7. The $^{R}Q_{2}$ region of v_{9} . (A) FTS spectrum; (B) diode laser scan; (C) deconvoluted diode data showing full resolution of the complex feature near 827.610 cm⁻¹; (D) simulated spectrum using full model.
- Figure 8. The $^{R}Q_{3}$ region of v_{9} . (A) FTS spectrum; (B) diode laser scan where the torsional splitting is only evident as shoulders; (C) deconvoluted diode laser data with torsional splitting now completely resolved even at J=4; (D) simulated spectrum using full model.
- Figure 9. The $^{R}Q_{4}$ region of v_{9} . (A) FTS spectrum; (B) diode laser spectrum;

- (C) deconvoluted diode data showing complete resolution of the torsional splittings even at J = 6; (D) simulated spectra using full model.
- Figure 10. The RQ_0 branch of v_9 . (A) FTS spectrum; (B) diode laser scan showing that even at Doppler limited resolution the fine structure is still unresolved; (C) deconvoluted diode data illustrating complete resolution of the unusual structure of RQ_0 . This is a result of a two-fold torsional effect for RQ_0 caused by the $3v_4$ resonance and the strong ℓ -doubling that is quite common in the RQ_0 branch of perpendicular bands of symmetric top molecules; (D) simulated spectra using the full model. Unassigned peaks are assumed to belong to the hot band $v_9 + v_4 v_4$.
- Figure 11. Effects of 2-resonance on the (A) $^RQ_1(J)$ branch and (B) $^PQ_1(J)$ branch of v_9 . The J values are numbered and both plots are on the same frequency and absolute intensity scales. Note the enhanced intensity of the $^RQ_1(J)$ lines relative to the $^PQ_1(J)$ with the same J value. The effect increases with J. A rigid rotor model would have the intensities approximately equal.
- Figure 12.A page from Part I of the ν_9 Band Atlas that covers 837.5 to 840.0 cm⁻¹. The upper trace in each row is the FTS data (the RQ_6 region in this case). The lower trace is the simulated spectrum using the full model. The torsional splittings are clearly seen for all transitions. The calculated resolution is 1.8 x 10^{-1} cm⁻¹ at T = 300° K (i.e., Doppler limited).

TABLE I. TORSIONAL SPLITTINGS IN v_9 OF $C_2^H_6$

K"	Int. (a)	Int. _{RATIO}	σ	Tor. Shift (b)
ν II – 2n	lavala all	1 1		
K even	levels, all	2:1	0	(2/3)S _{0.9}
	W		2	-(1/3)S _{0.9}
K odd	S	2:1	3	-(2/3)S _{0.9}
	W		1	$(1/3)S_{0,9}$
<u>K"</u> ≠ 3p	levels, all	J		
K even	S	4:1	2	-(1/3)S _{0,9}
	W		0	(2/3)S _{0.9}
K odd	S	4:1	1	(1/3)S _{0,9}
	W	1.4	3	-(2/3)S _{0,9}
K" = 0 1	<u>evel</u>			
J even	S	6:2	0	(2/3)S _{0.9}
	W	V , L	2	-(1/3)S _{0,9}
J odd	S	10:6	0	(2/3)S _{0.9}
	W	10.0	2	-(1/3)S _{0,9}

 $⁽a)_S = stronger and W = weaker component$

⁽b) $S_{0,9} \equiv |\Delta_9 - \Delta_G|$ is the difference in torsional level splittings between the ground and v_9 vibrational states.

TABLE II. WHOLE BAND ANALYSIS OF v_9 OF $C_2H_6^{\dagger}$

	Present Study	Daunt, et al. (a)
v ₀	821.7234	821.727(6)
Α α ₉	$-7.90722(59)\times10^{-3}$	$-7.837(30) \times 10^{-3}$
Β α ₉	$1.27454(13)\times10^{-3}$	1.259(15)x10 ⁻³
Ας Z	0.694742(13)	0.6950(5)
ე ო <u>ფ</u>	-2.410(28)x10 ⁻⁶	-2.42(1.30)×10 ⁻⁶
K ng	3.0758(49)×10 ⁻⁵	$3.448(1.300) \times 10^{-5}$
β ₉	-1.66(13)x10 ⁻⁹	ND(c)
β <mark>JK</mark>	-5.58(12)x10 ⁻⁸	ND
β <mark>K</mark> β9	$-1.810(38)\times10^{-7}$	ND
q_9^+	-1.8169(14)×10 ⁻³	$-1.832(30)\times10^{-3}$
ر 69ۇ	1.98(16)x10 ⁻⁸	ND
δq <mark>K</mark>	$-8.80(33)\times10^{-7}$	ND
σ(S.D.)	0.7×10^{-3}	6.7x10 ⁻³
N/N ₀ (b)	1342/1609	549/569

 $^{^{\}dagger}$ All quantities in units of cm $^{-1}$; error limits are 95% confidence limits.

⁽a) Daunt, S.J., Blass, W.E., Halsey, G.W., Fox K. and Lovell, R.J., 1981, J. Mol. Spectrosc. <u>86</u>, 327.

 $^{^{(}b)}N/N_0$ is the ratio of lines included without zero weight (N) to the total number of assigned lines (N_0) .

⁽c)_{Not determined.}

TABLE III. COMPARISON OF RECENT DETERMINATIONS OF THE GROUND STATE CONSTANTS FOR ETHANE[†]

	PRESENT WORK	PINE AND LAFFERTY (a)	LIN, ET AL. (b)	COLE, ET AL. (c)
8 0	0.6630279(24)	0.6630271(14)	0.6630353(152)	0.663089(24)
$D_0^{J}(\times 10^{-6})$	1.0324(23)	1.0312(26)	1.0406(16)	1.084(35)
$D_0^{JK}(x10^{-6})$ 2.651(88)	2.651(88)	2.660(29)	2.575(274)	5.00(66)
$\sigma(x10^{-3})$	0.87	0.55	2.2	ı
(P) ⁰ N/N	760/893	(e)	230/285	184(e)

[†]All quantities are in units of cm⁻¹.

⁽a)Pine, A.S. and Lafferty, W.J., 1982, J. Res. Natl. Bur. Stand., 87 237; uncertainties are one standard deviation.

⁽b)Lin, K.F., Blass, W.E. and Gailar, N.M., 1980, J. Mol. Spectrosc. 79, 151.

⁽c)Cole, A.R.H., Cross, K.J., Cugley, J.A. and Heise, H.M., 1980, J. Mol. Spectrosc. 233; uncertainties are two standard deviations.

⁽d)See N/N_D comment in Table II.

⁽e)These calculations performed with standard fixed weighting methods.

TABLE IV. INTENSITY EFFECTS OF &-RESONANCE

	<u>K"</u>	<u>j"</u>	Relative ^a Increase in R-type (ΔK=+1) Transitions	Relative ^b Decrease in P-type (ΔK=-1) Transitions	Percent Intensity Over(under) estima- tion for R(P) Transitions
•					
	1	10	1.048	0.962	4
	ī	15	1.084	0.917	4 8 15 22
D	ĩ	20	1.145	0.855	15
		25	1.221	0.779	22
	ī	30	1.309	0.691	31
	1	35	1.403	0.597	40
	1 1 1	40	1.497	0.503	50
	2	10	1.018	0.982	2 7
	2	15	1.072	0.928	7
	2 2 2 2 2	20	1.112	0.888	11
	2	25	1.160	0.840	16
	2	30	1.215	0.785	21
	3	10	1.012	0.988	1
	3 3 3	20	1.048	0.952	1 5
	3	30	1.107	0.893	10
	4	10	1.008	0.992	1
	4	20	1.035	0.965	1 3 8
	4	30	1.080	0.920	8
	5	10	1.006	0.994	0
	5 5 5	20	1.028	0.972	0 3 6
	5	30	1.063	0.937	6

a Intensity ratio of R(perturbed)/R(unperturbed)

b Intensity ratio of P(perturbed)/P(unperturbed)

TABLE V. LIST OF $^{14}\mathrm{Co}_2$ LASER LINES AND NEARBY COINCIDENCES WITH $^{92}\mathrm{of}$ $^{22}\mathrm{H}_6$ TRANSITIONS

	Relative Intensity		1.50784 0.37696 0.25736 0.30736 0.36432 0.42872	•		0.79653 0.35484	0.39827 1.41936 0.65653	1.96958	0.37214 1.48856	0.30130 1.20520	1.54256 0.39448	1.57792 0.36416	1.45664 0.39772	1.59088 0.39442	1.57768	0.41706	1.53464 0.36448
	Assignment AKAJ(K"J"g")		RR(2,2,2) RR(2,2,0) RQ(4,33,2) RQ(4,32,2) RQ(4,31,2) RQ(4,30,2)			(3,15, (5,18,	 6	(0,9,0 (2,13)	(5,17,5)		(5,16, (5,15,	(5, 15, 11, 13, 13, 13, 13, 13, 13, 13, 13, 13	1,7,1 5,14,	5,14, 5,13,	5,13,	2,5,0 5,12,	
-) _{\Dv} (MHz)		-5224 -5156 -4428 -2019 324 2602 4815			-5445 -5387	-5315 -5281 -5129	-5062 -4564	-3996 -3895	-3535 -3436 -2680	-2583 -1438	-1344 -550	-471 -272	-183 815	901 1229	1305 1829	1911 2765
	°CO ₂ (cm ⁻¹)	831.81368	831.63944 .64171 .66598 .74632 .82449 .90048	833.68367	835.53734	835.35570	.36005 .36119 .36626	.36848	7	777	.45119 .48939	. 51901	.52163	.53123	.56740	.58087	.62957
	CO ₂ Laser	P(40)		P(38)	P(36)												
	Relative <u>Intensity</u>		0.70824 0.81464 0.92992 0.26334 1.05336 0.29600	-, (,) (,	10,4		0.26504 0.39225				0.71817 1.55853						
	Assignment ΔΚΔJ(Κ"J"σ")		RQ(1,29,1) RQ(1,28,1) RQ(1,27,1) RQ(1,26,3) RQ(1,26,1) RQ(1,26,1)	(1,25)	(1,23)		RQ(2,34,2) RR(0,2,0)			(3,22, (3,21,	RQ(3,21,1) RQ(3,20,3)	(3, 50, (3, 19,	, 6, 6, 6, 6, 6, 6, 6, 6, 6, 6, 6, 6, 6,	(3, 18, 17, 17, 17, 17, 17, 17, 17, 17, 17, 17	(3,1/, (3,16,	(3,15,1 (3,15,3	3,14,
•	Δν(MHz) [†]		-4648 -2868 -1122 394 583	2250 3703 3874	5289 5451		4542 5965			-5903 -4401	-4268 -2834	-1331	97	1457	2743	2848 3958 4058	4036 5101
	$^{\text{CO}_{2}}_{\text{C}_{2}\text{H}_{6}}$	824.16974	824.01469 .07408 .13232 .18289 .18919	. 29327 . 29327 . 29897	.34615	826,10540	826.25690 .30437	828.02458	829.92733	829.73042 .78052	.78497 .83281 .83705	.88293	.93057	.97594	830.01883	.02233	.09749
	CO ₂ Laser	P(48)				P(46)	- 44: -	P(44)	P(42)								

	1.06952 0.25704 1.02816 0.95792	.8548 .7140 5300		.2876	0.28312 0.28312 0.62275	.8682 .6052		.2679	.3364	1.66984	.41/t		.4805	5332	0.26190	. 2822 . 2822	. 5922 . 3688	6009	. 5004 . 6312	3156	3265	3321	
	14 13 12	(7,11,1 (7,10,1)		(5,25,	RQ(8,29,2) RQ(8,29,2) RR(0,15,2)	(0,15,		(1,18,	(1,18 (1,18	RR(2,12,2) RR(2,12,2) RR(2,12,0)	, 21, 2)		(9,23,	4,0,0	RQ(9,22,3) RQ(9,22,1)	(9,71,	(5,7,1	20,00	9,20,	ο ο σ		9,17,	
	998 2020 2101 3130	07 94 74		02	4621 4621 4993	06 84		544	30 64 77	3406 3518	7		505	378	-3223	136 136	14 -7	\circ	36	33	1 Ö 1	ر 2 ک	
	.03395 .06806 .07075	66 57 21	7893	8901	.93116 .94345 .95586	9580 9843	844.56172	844.38000	ין עון ע	. 67534 . 67534		846.31794	846.14949	1516	.21402	.27232	.31549	32	.37794	.38112	. 43161	.47952	
V CONTID)			P(28)				P(26)				•	P(24)											
(TABIF	45792 97080 98540 33594	. 343 . 296 . 187	.984		.2850	0.39147	.2604 .5209		.2571	0.53712 0.50554	.8425	3647	.9551	.2246		•		•	• •	•	•	• •	0.27146 1.08584 0.26738
	RQ(5,11,1) RR(3,3,3) RR(3,3,1) RQ(5,10,3)	(5, 10, 10, 10, 10, 10, 10, 10, 10, 10, 10	(5,8,1 (5,7,1		0(6,31,0	RQ(6,29,0) RQ(6,28,0)	0(6,27,0		(6,24, (5,22,	PR(4,20,2) RR(0,12,2)	(0, 12, 0)	(3, 18) (3, 18)	(2,16,116,111,111,111,111,111,111,111,111	(1, 14,		(7,19,	(7, 18, 1)	(3,7,3)	(7,17,	(',1',1 (7,16,3	(7,16,1	(4,5,0 (4,5,0	RQ(7,15,3) RQ(7,15,1) pn/7,14,3
	2844 3213 3281 3624	568	18 80		99	1612 3813	94		55	-1427 -412	34	34 18	03 52	63		568	419	62 55	287	4/5 153	144	65	-272 -186 914
	. 63222 . 64452 . 64677 . 65822	843 868	7101	837.37471	746 527	. 50189	5729 5729	839,19581	1104 1234	.14821	1842	1843 1896	2304 2798	2837	841.00067	ī	. 0	$\sigma \sim$	904	> ດ	952	യ	. 99158 . 99448 . 03114

P(34)

6 - 45 -

P(30)

P(22)	848.05797			(TABLE V.,	, CONT'D.)				
	847.92096 .92318 .92795 848.00364 .08787	-4107 -4041 -3898 -1629 896 3433	RR(0,19,2) RR(0,19,0) PR(4,27,2) PR(3,25,3) PR(2,23,2) PR(1,21,1)	0.48790 1.46370 0.27408 0.41340 0.60376 0.87912		.32425 .42528 .47923 .48383 .64585	-5732 -2703 -1086 -948 3909 4007	RR(3,19,1) RR(0,26,0) RR(4,17,2) RR(4,17,0) RR(5,15,3) RR(5,15,3)	0.56583 0.28556 1.39280 0.34820 0.40944 1.63776
P(20)	849.78182				P(10)	858.15839			
	849.70794 .71233 .81382 .81866 .93555	-2215 -2083 -2083 1104 4609 4709	RR(1,18,3) RR(1,18,1) RR(2,16,2) RR(2,16,0) RR(3,14,3) RR(3,14,3)	0.25402 1.01608 1.35968 0.33992 1.65467		858.09157 .10651 .10996 .29436	-2003 -1555 -1452 4076 4157	PR(1,29,1) RR(6,14,2) RR(6,14,0) RR(7,12,3) RR(7,12,1)	0.36008 0.86760 1.73520 0.47878 1.91512
p(18)	851 48950				P(8)	859.78513			
- 46	51.35 51.35 .36 .50	-3973 -3865 385	RR(4,13,2) RR(4,13,0) RR(5,11,3) PB(5,11,3)	1.82808 0.45702 0.51668		859.78340 .78606 .85442	-52 28 2077 5294	RR(8,11,2) RR(8,11,0) RR(1,26,1) RR(2,24,2)	1.93760 0.48440 0.37184 0.59776
5. -	. 59406 . 59406	46/ 3135 3201	(0,22,0)		P(6)	861,39566			
	.55525 .65717 .65981	5201 5027 5106	(6,9,2) (6,9,0) (6,9,0)			21 28 28	-5316	RR(2,25,2) RR(9,10,3)	.9218
P(16)	853.18100					ვ ფ	321 -56 -41	(3,73, 2,33,40,40,40,40,40,40,40,40,40,40,40,40,40,	0.72447 0.36223
	853.07035	-3317	2,27,	.382		45	79	(4,21,	.9406
	.12122	-1792	RR(7,8,1) PR(1,25,1)	2.35920 0.59968	P(4)	862,98995			
P(14)	854.85631	į				862.81721 863.00960 01367	-5179 589 711	RR(4,22,2) RR(5,20,3) RR(5,20,1)	0.8368E 0.26128
	854.80056	-1671	1,22	0099.		,0010.	77,	(0,50)	ا :
	.81380 .86813	354	12,2 12,1	.2573 .2610	† The con	conversion from wav	wavenumber	to frequency wa	s calcula
	.90904 .91979 .855.04873 .05276	1581 1903 5769 5889	KK(2,20,2) RQ(12,18,0) RR(3,18,3) RR(3,18,1)	0.98312 0.25820 1.24067 0.62033	with a	value of c = 2.9	9792458	x 10 ¹⁰ cm/sec.	
P(12)	856.51545								

	0001469999999999999999999999999999999999	
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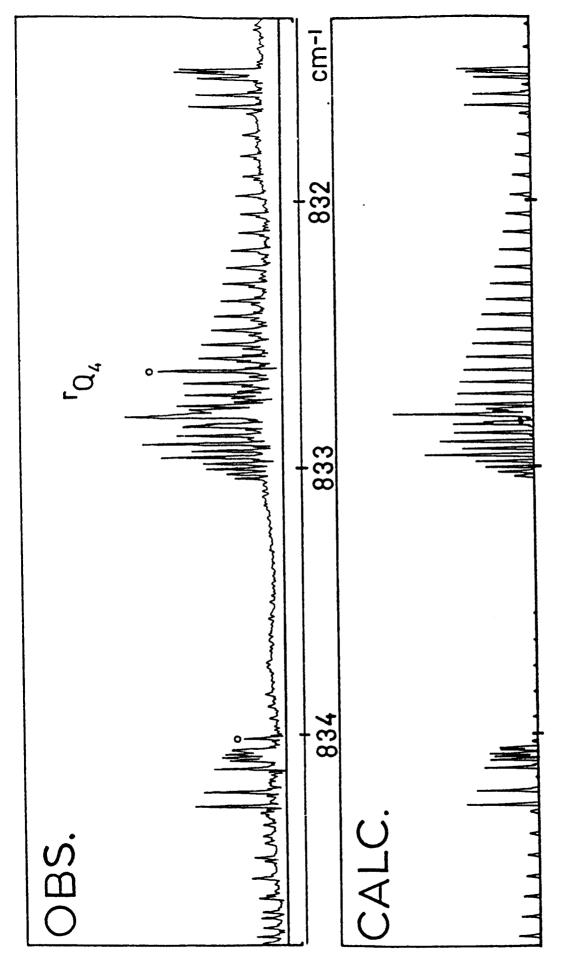
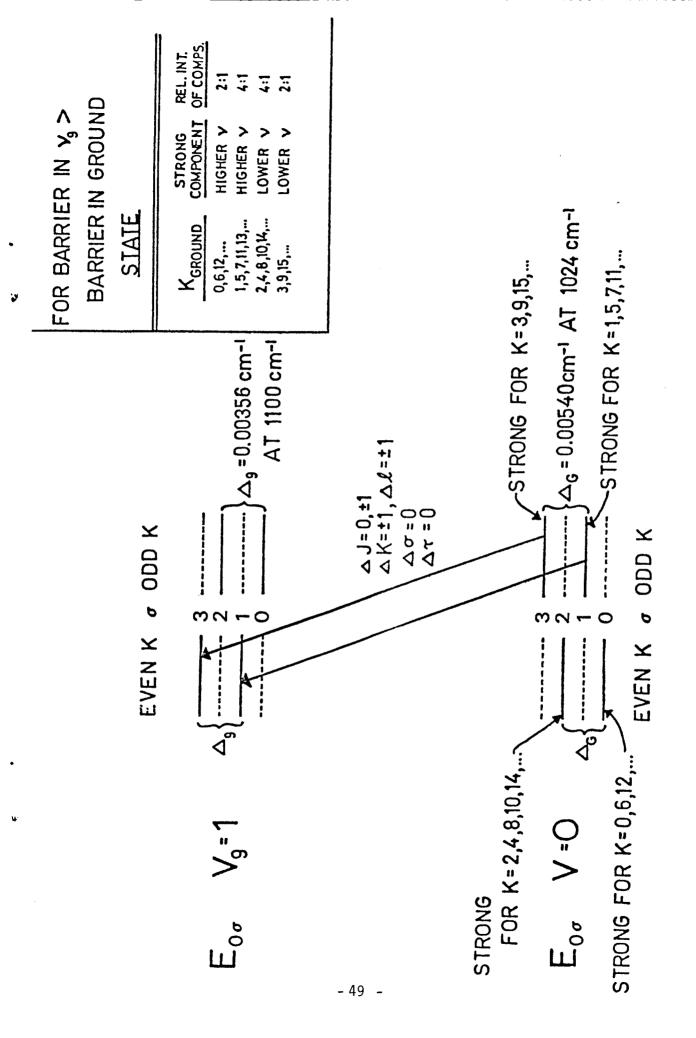


Fig. I Dant et al



2.8.2 Daunt etal.

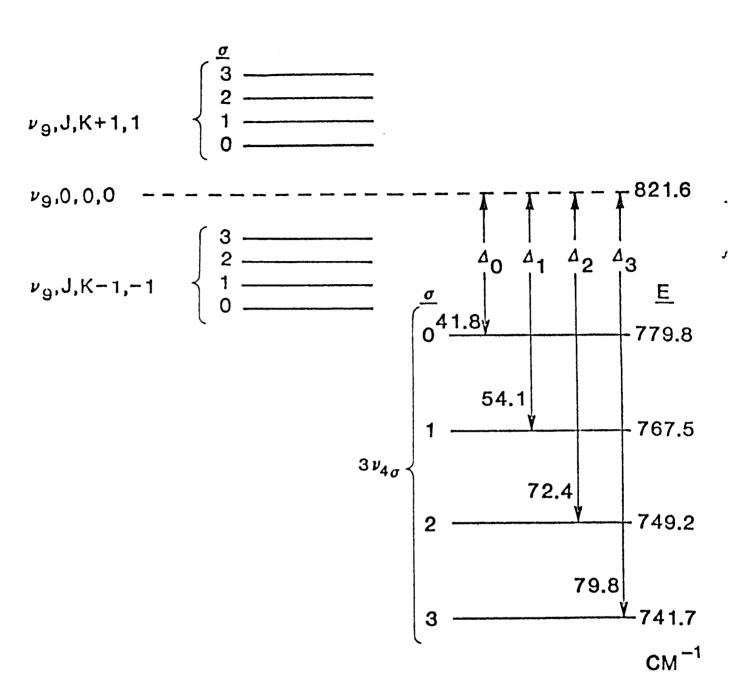
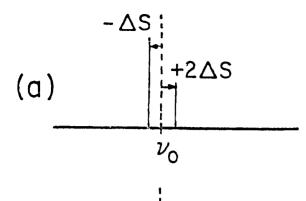
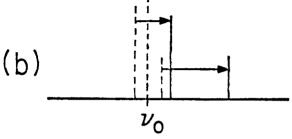


Fig. 4 Daunt et al.



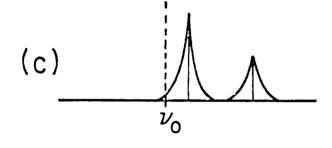


Torsion only

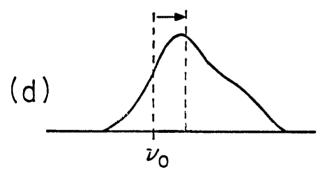
[Total splitting =
$$3\Delta S = S_{0,9}$$

= $2.2 \times 10^{-3} \text{ cm}^{-1}$]

+ Resonance with $3\nu_4$



- (1) Doppler line shape
- (2) Add



- (1) Convolute with instrument function
- (2) Find maximum (a)
- (3) Calculate shift (s) from ν_0
- (e) Merge measurements with shifts
- (f) Use standard model +l resonance

Fig. 5 Dauntet al.

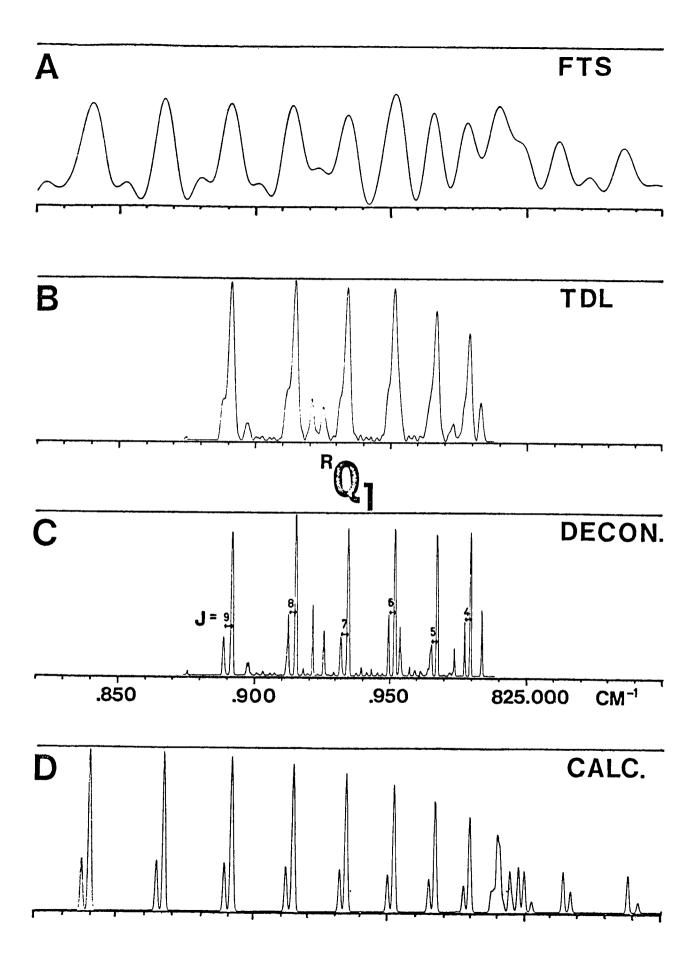


Fig. 6 Daunt et al.

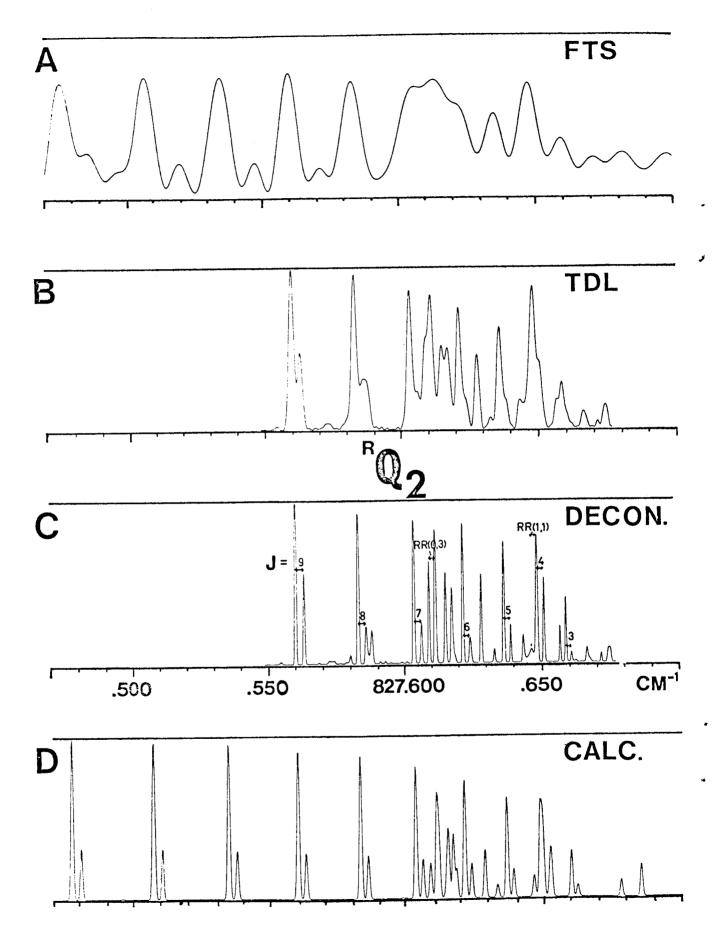


Fig. 7 Daunt et al.

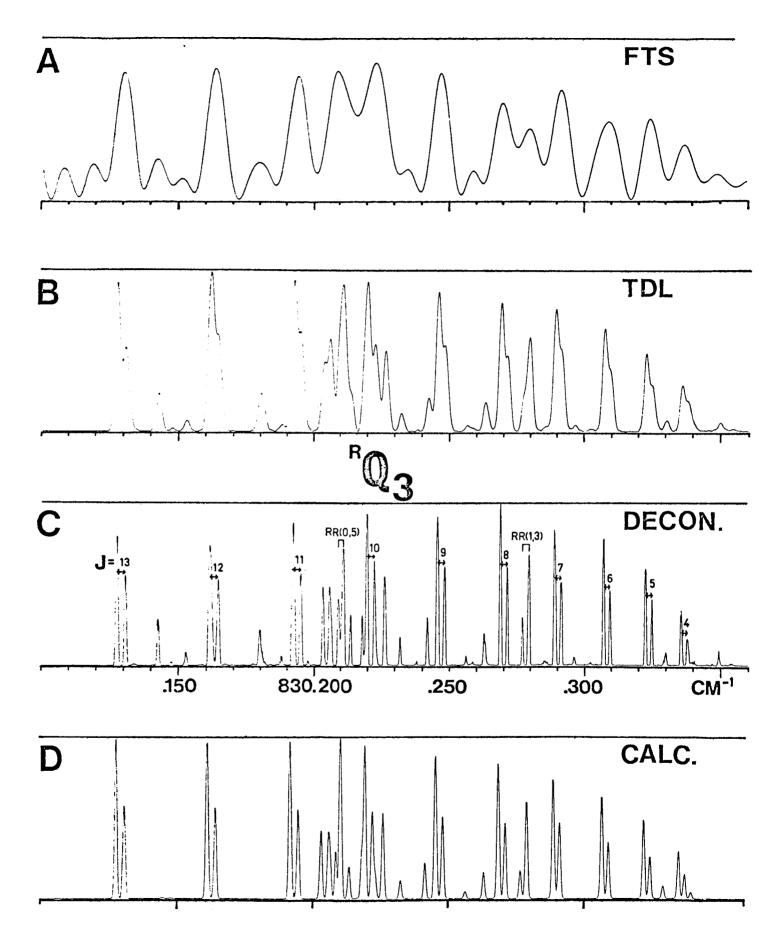


Fig. 8 Dauxt et al.

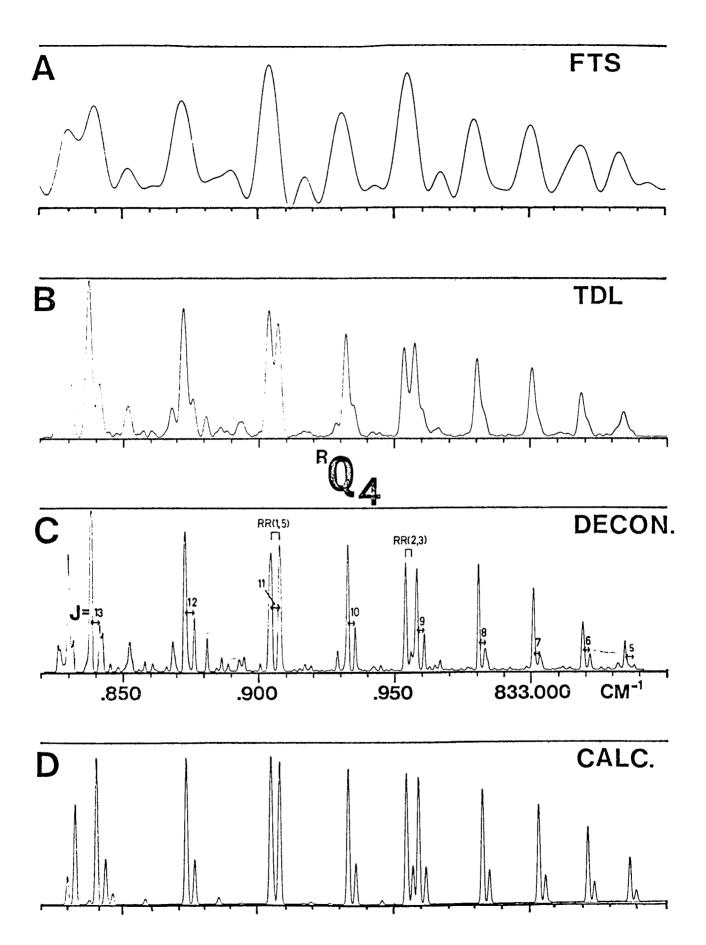


Fig. 9 Daunt et al.

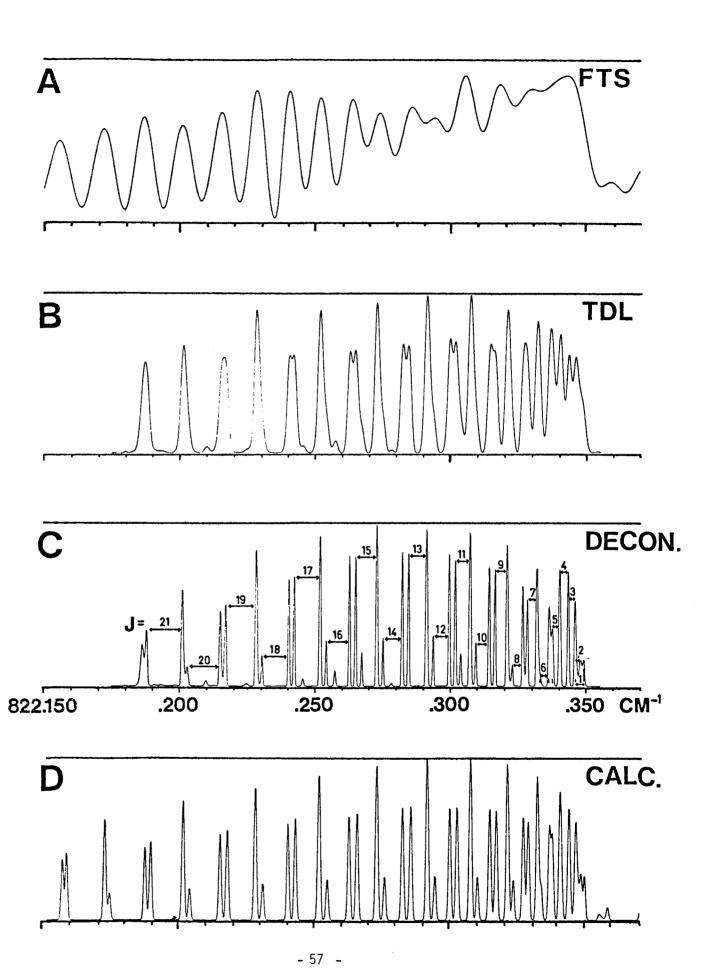
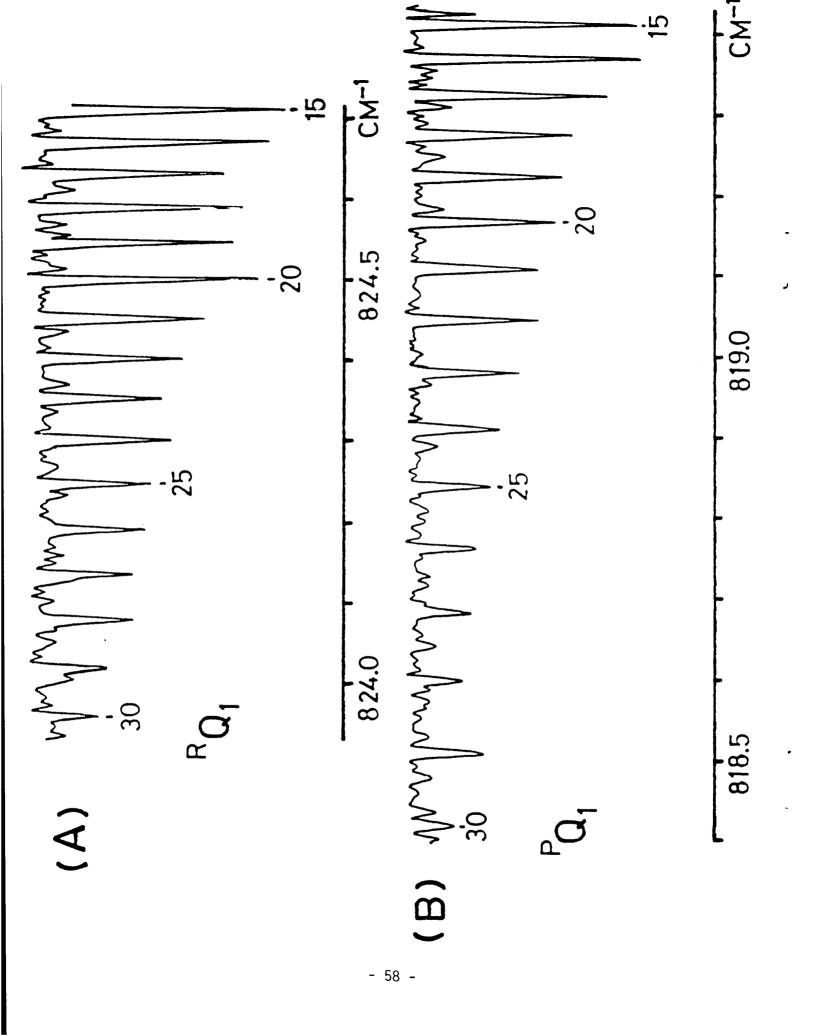
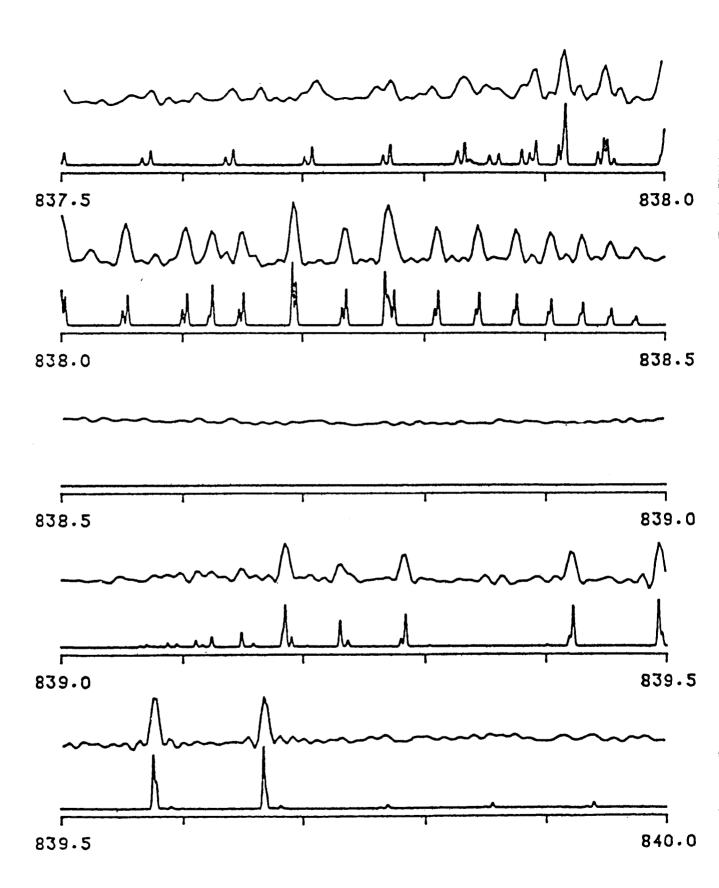


Fig. 10 Dauxt et al.





PART II

- 62 -

 $${\rm The}\ 12\mu m\ Band\ of\ Ethane}$$ A Spectral Catalog from 765 ${\rm cm}^{-1}$ to 900 ${\rm cm}^{-1}$

A.K. Atakan, W.E. Blass, S.J. Daunt and G.W. Halsey
The University of Tennessee
Molecular Spectroscopy Laboratory

D.E. Jennings, D.C. Reuter and J. Susskind NASA/Goddard Space Flight Center

and

J.W. Brault Kitt Peak National Observatory

Prepared

: Under NASA Contract #NAS5-26896 to The University of Tennessee

Ву

: A.K. Atakan and W.E. Blass

Principal Investigators

: W.E. Blass and S.J. Daunt

December 1982

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Figure 1

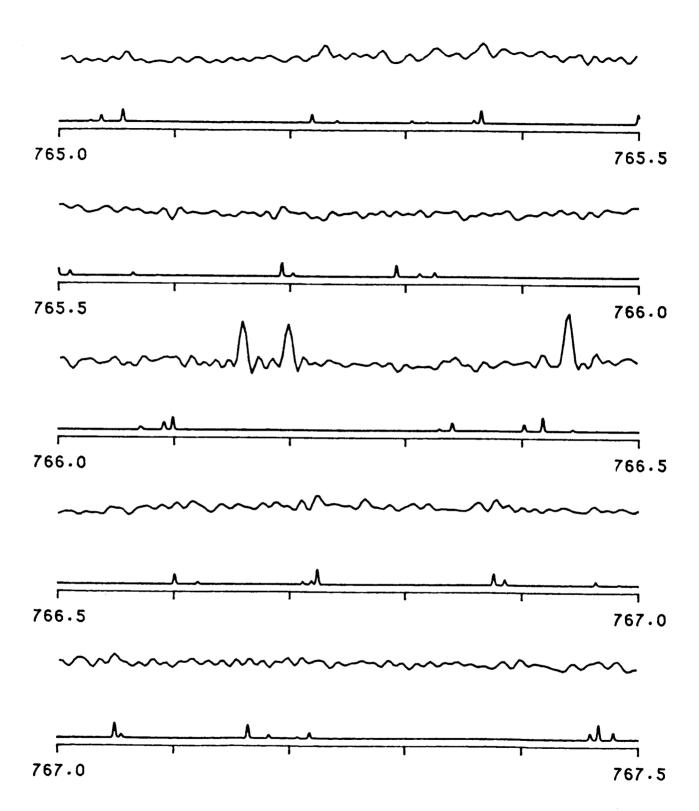
The observed $0.005~\rm cm^{-1}$ resolution spectrum taken at KPNO on the McMath Interferometer is presented in the upper trace of each panel, the calculated spectrum[†] (at an effective resolution of $0.0018~\rm cm^{-1}$) is presented in the lower trace of each panel. The relative intensities correspond to 300° K ethane and the apparent line width corresponds to an infinitely high resolution spectrum of Doppler limited 300° K ethane.

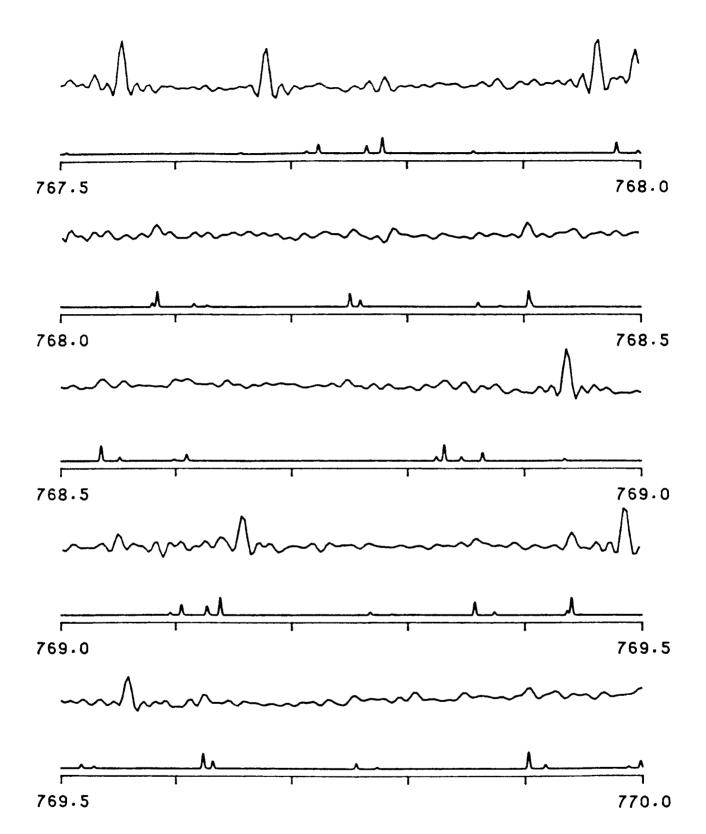
[†]The calculated spectrum is based upon the results of:

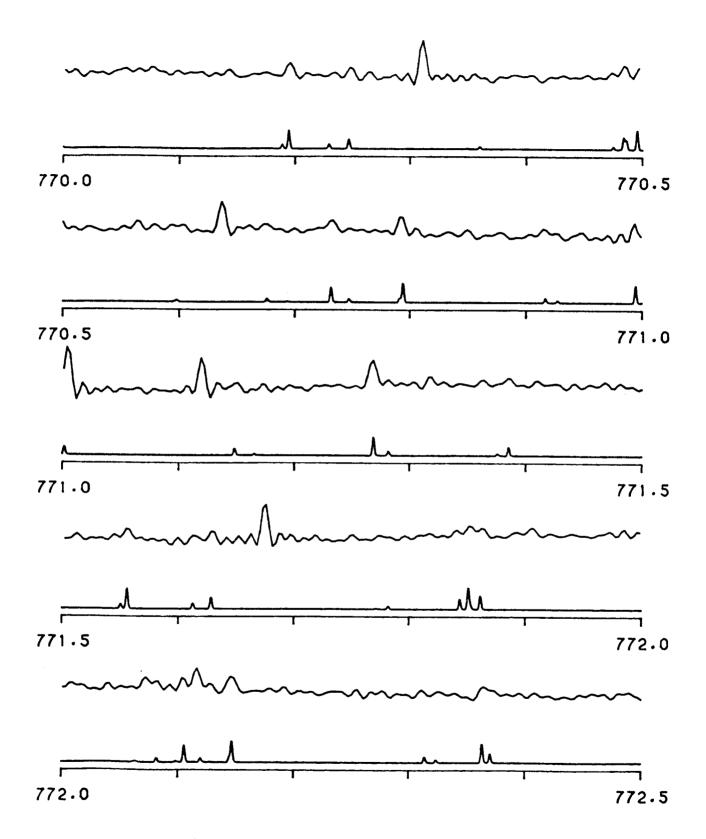
^{(1) &}quot;Diode laser spectra of the torsional splittings in the v_9 band of ethane: Torsion-vibration-rotation interactions and the barrier to internal rotation", J. Susskind, D.C. Reuter, D.E. Jennings, S.J. Daunt, W.E. Blass and G.W. Halsey, J. Chem. Phys., 77, 2728 (1982).

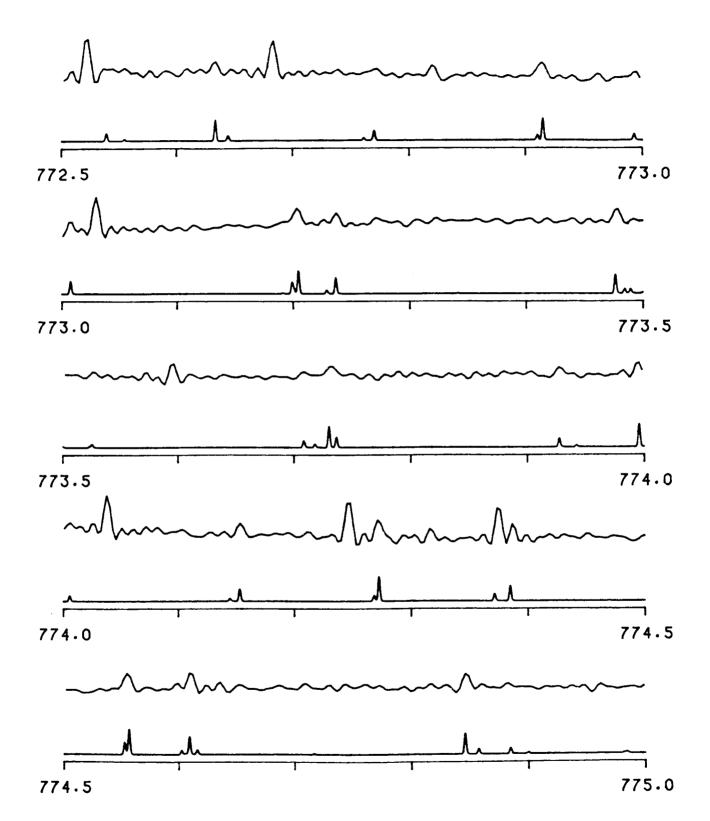
^{(2) &}quot;The 12µm Band of Ethane: High Resolution Laboratory Analysis with Candidate Lines for Infrared Heterodyne Searches", S.J. Daunt, A.K. Atakan, W.E. Blass, G.W. Halsey, D.E. Jennings, D.C. Reuter, J. Susskind and J.W. Brault, submitted to Ap. J.; Part I of this memorandum.

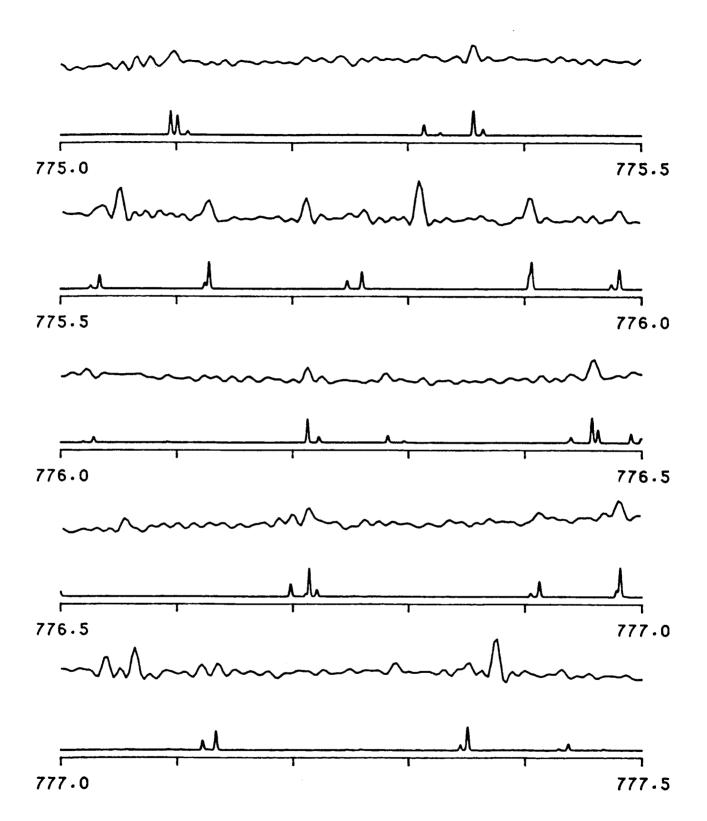
- 66 -

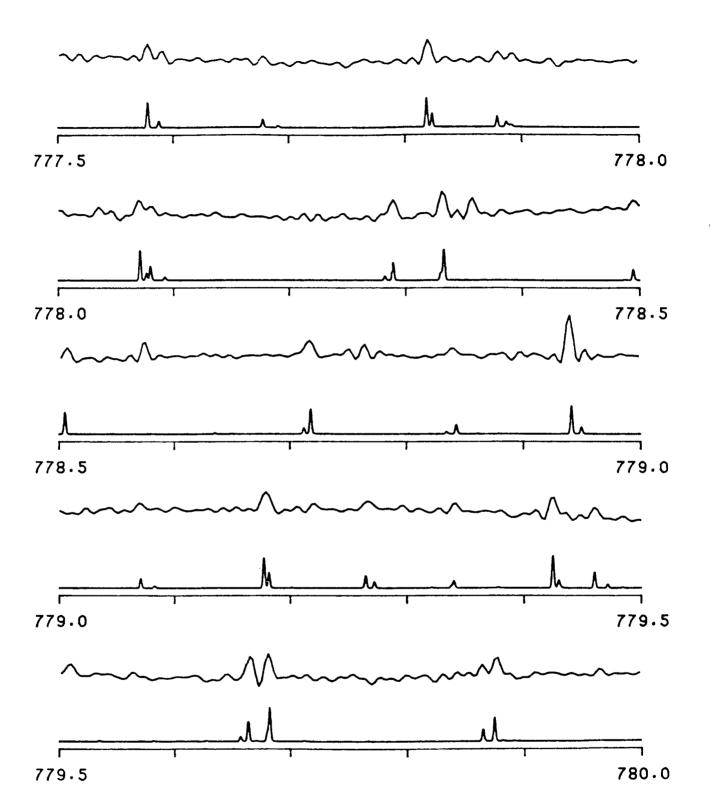


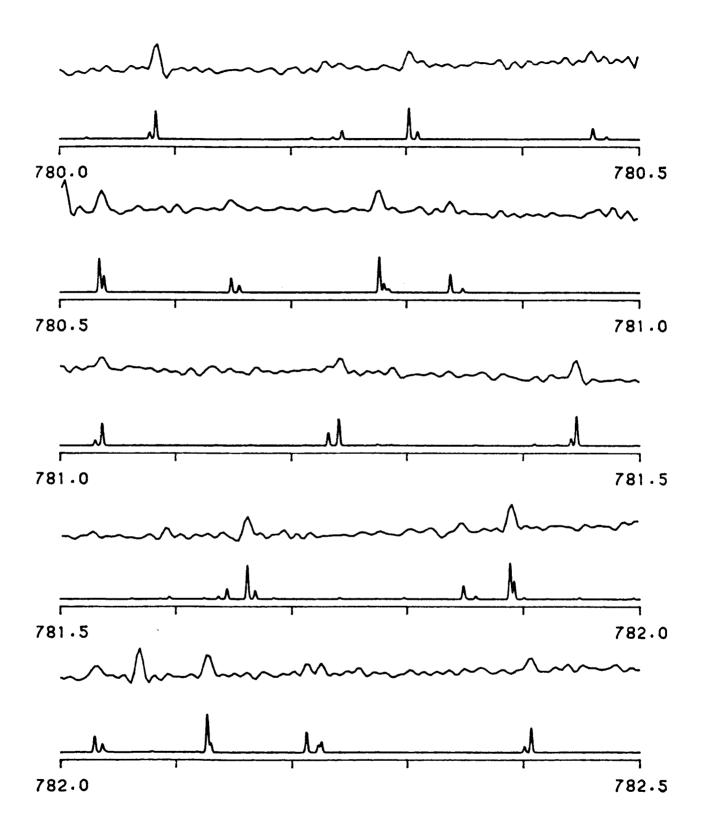


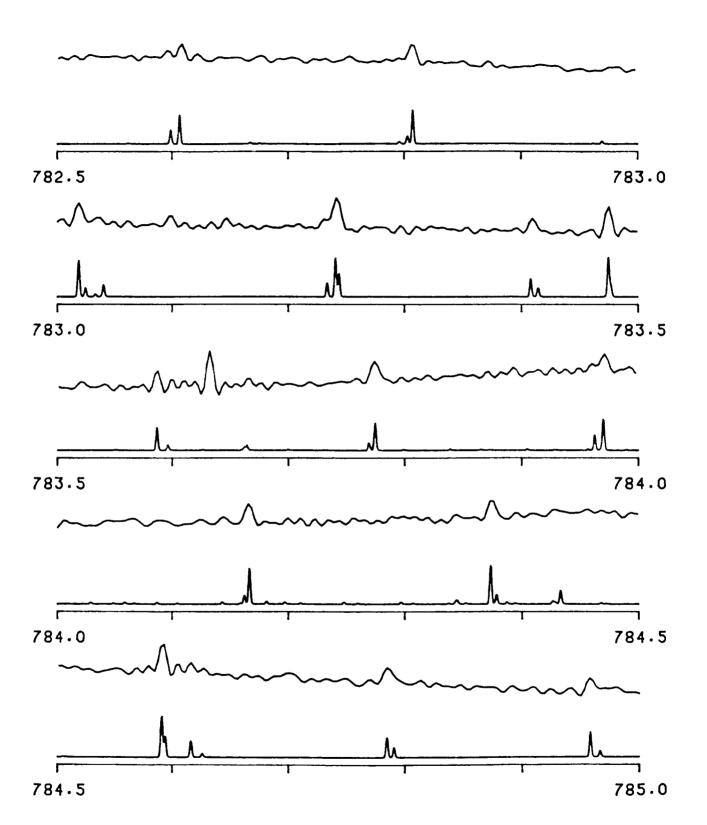


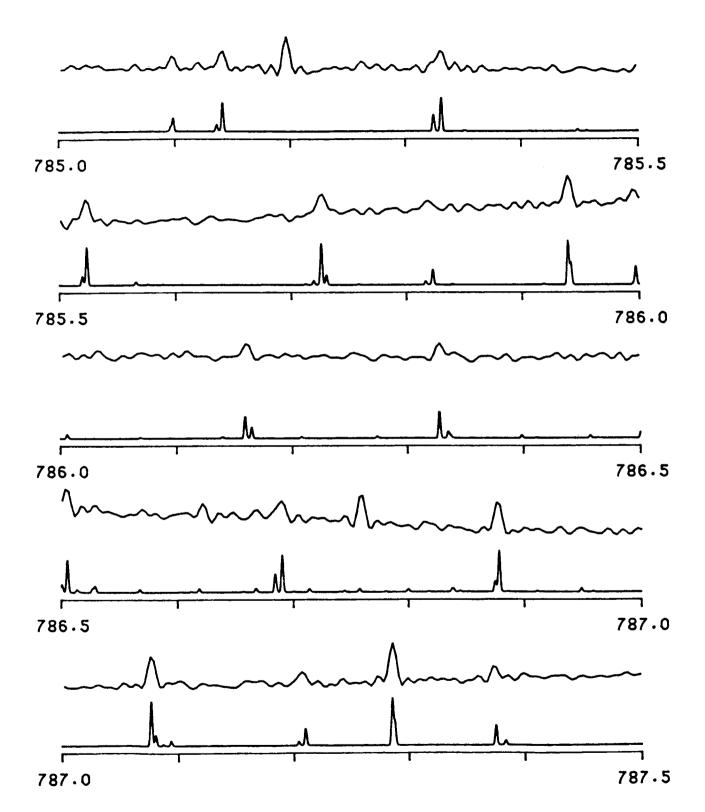


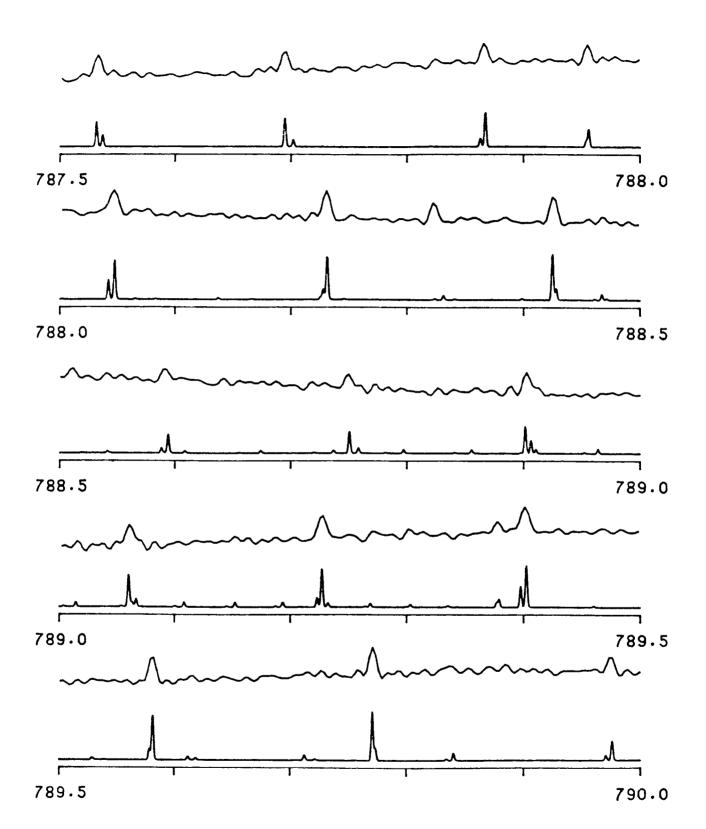


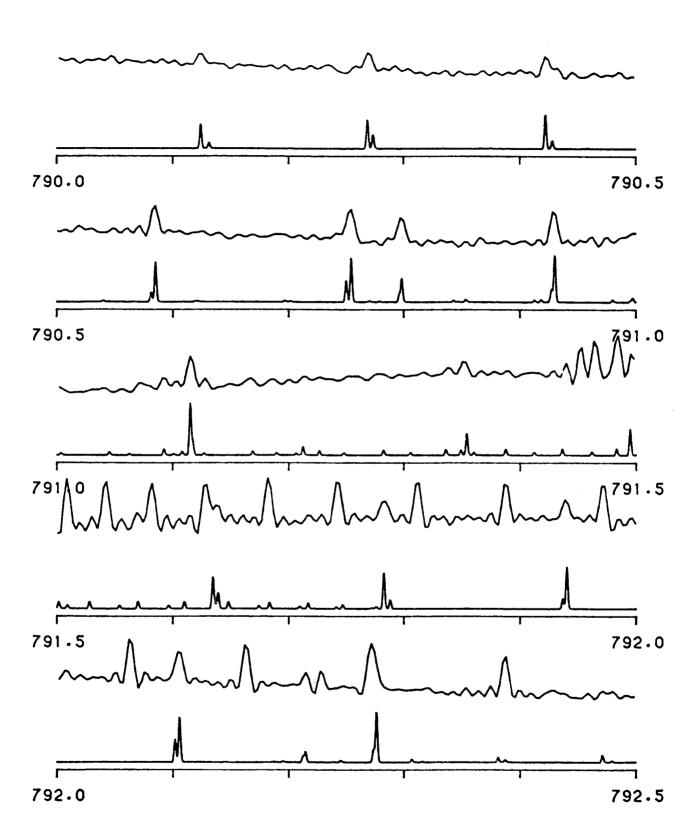


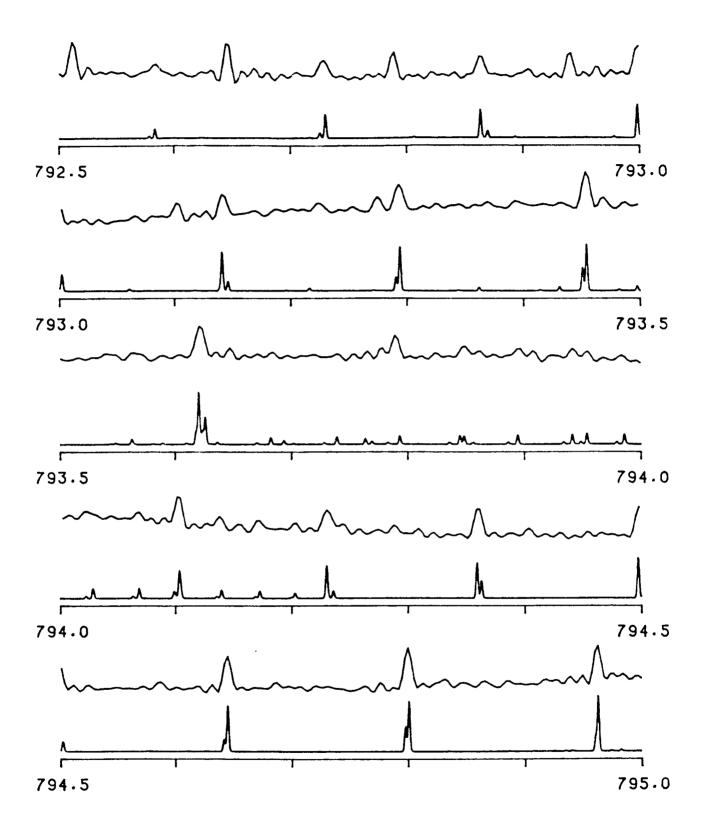


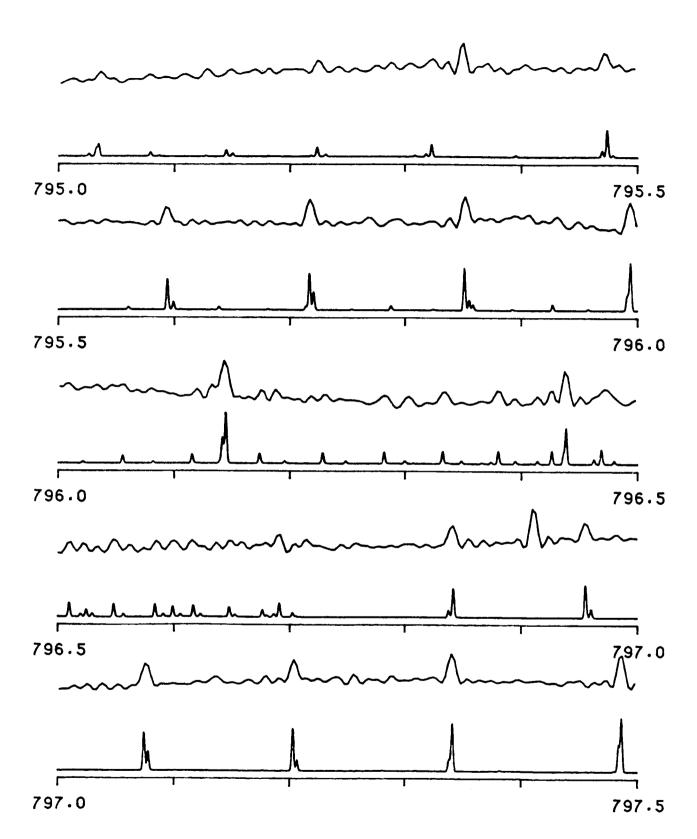


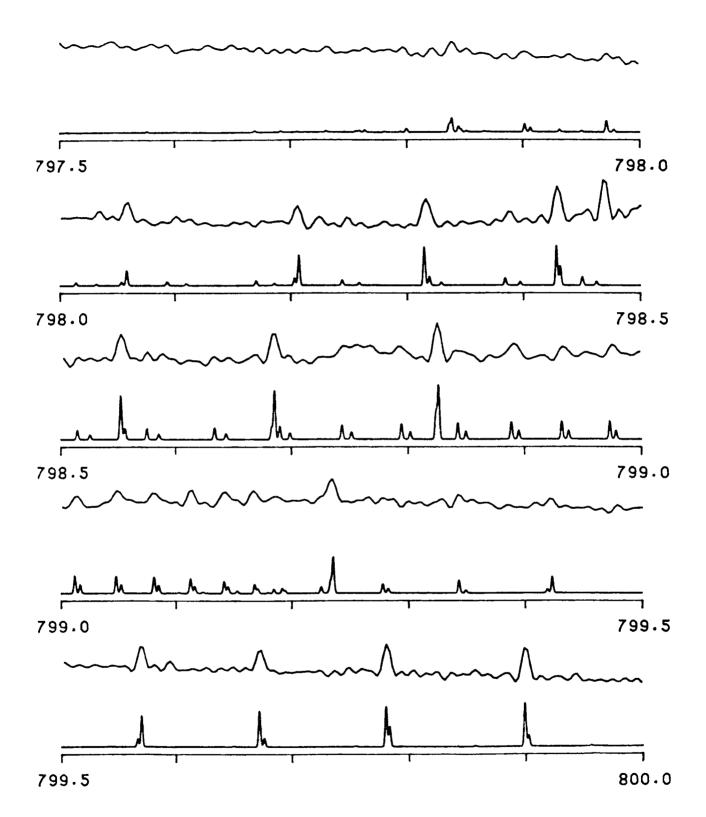


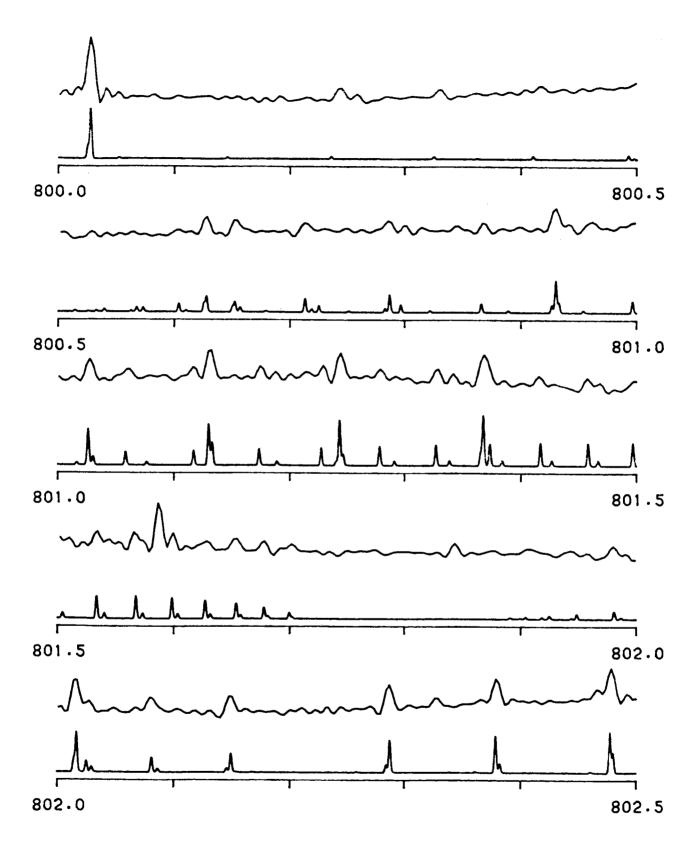


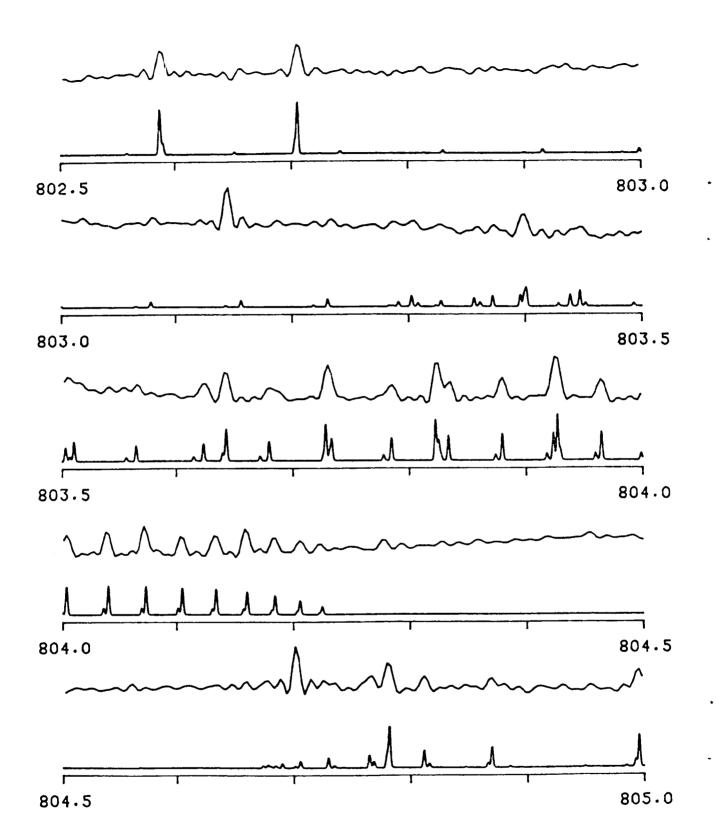


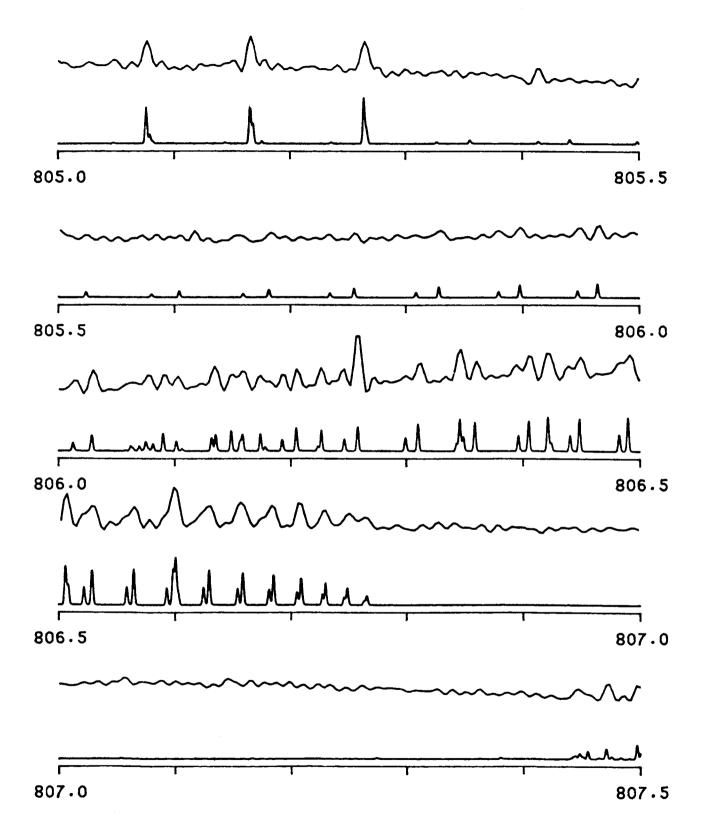


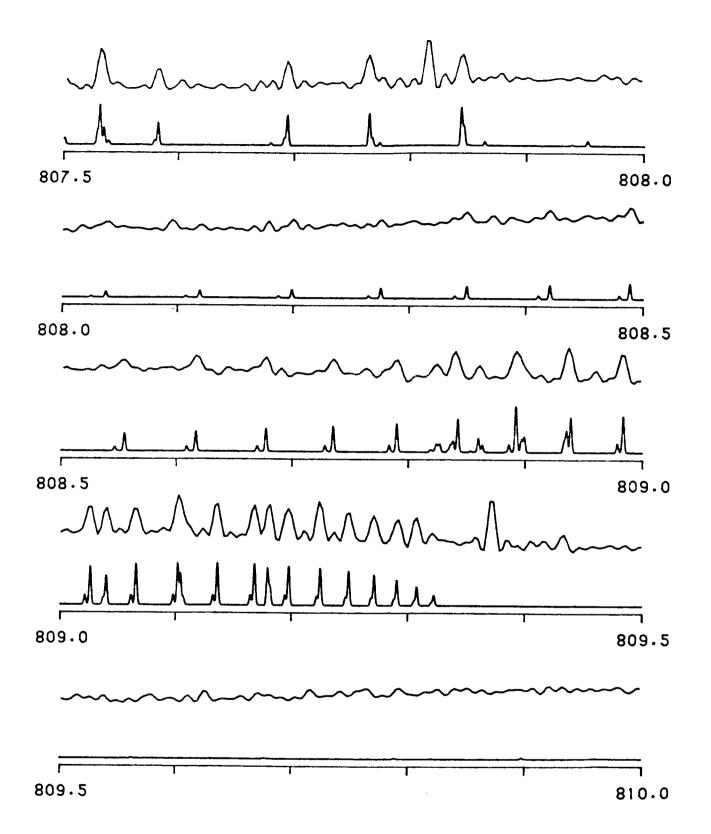


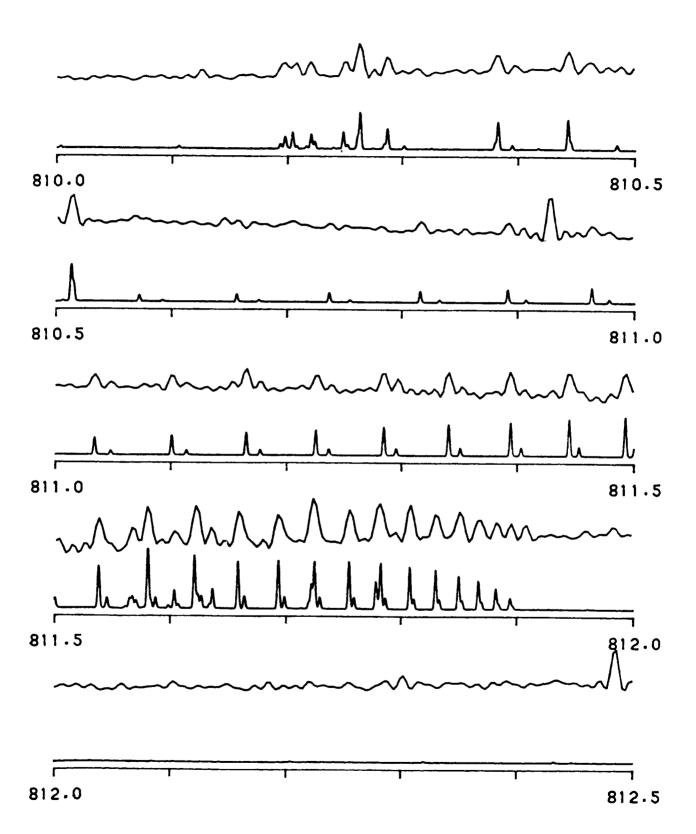


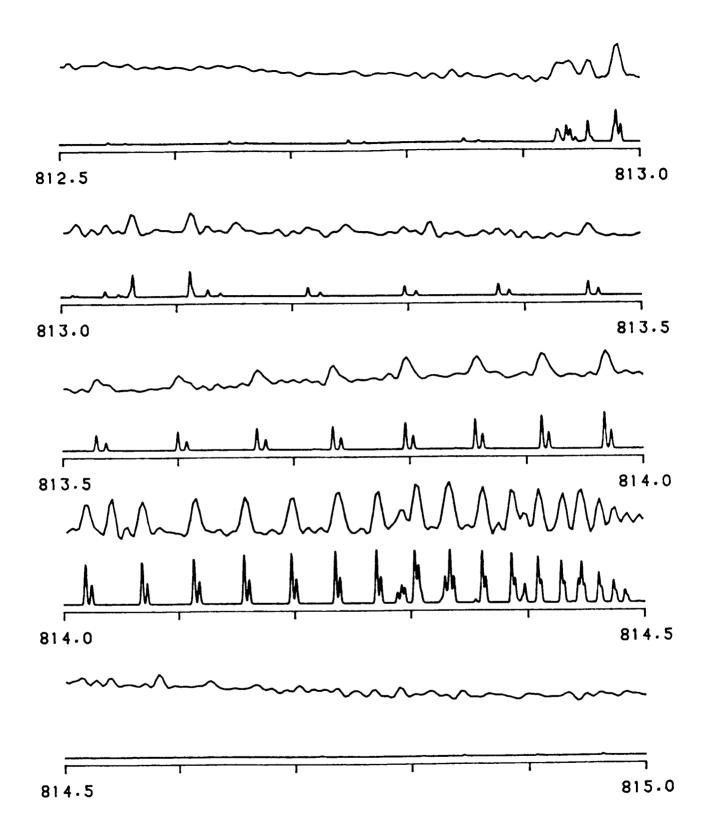


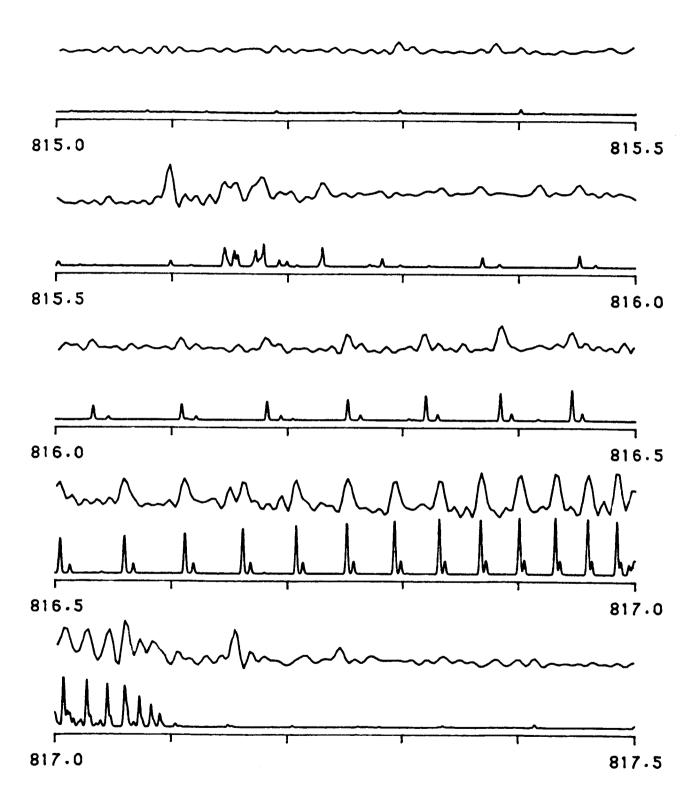


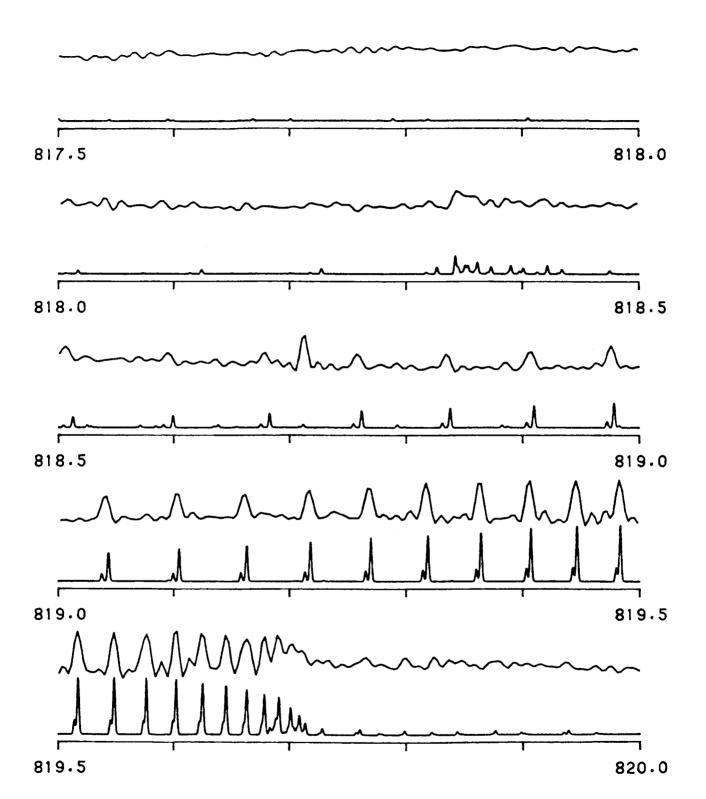


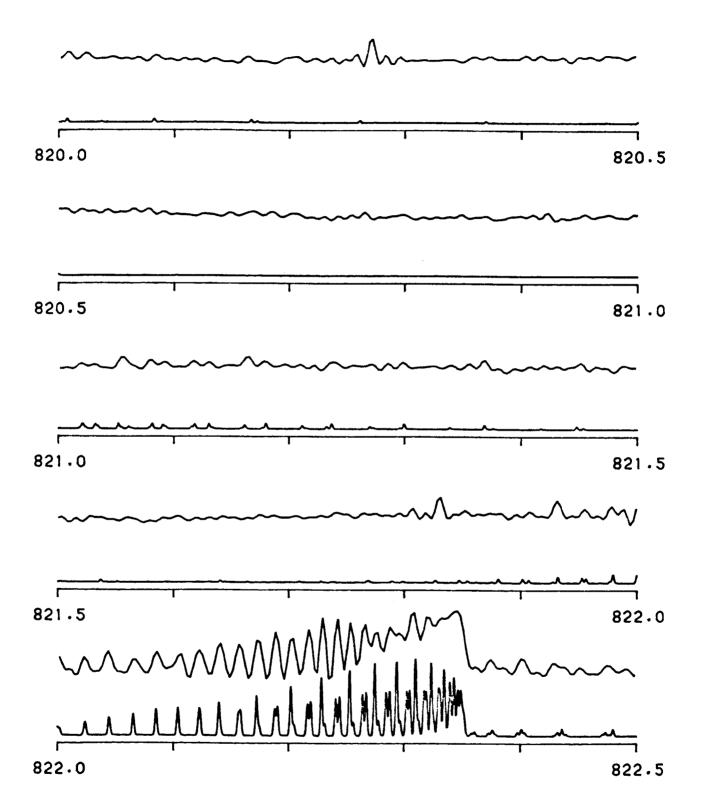


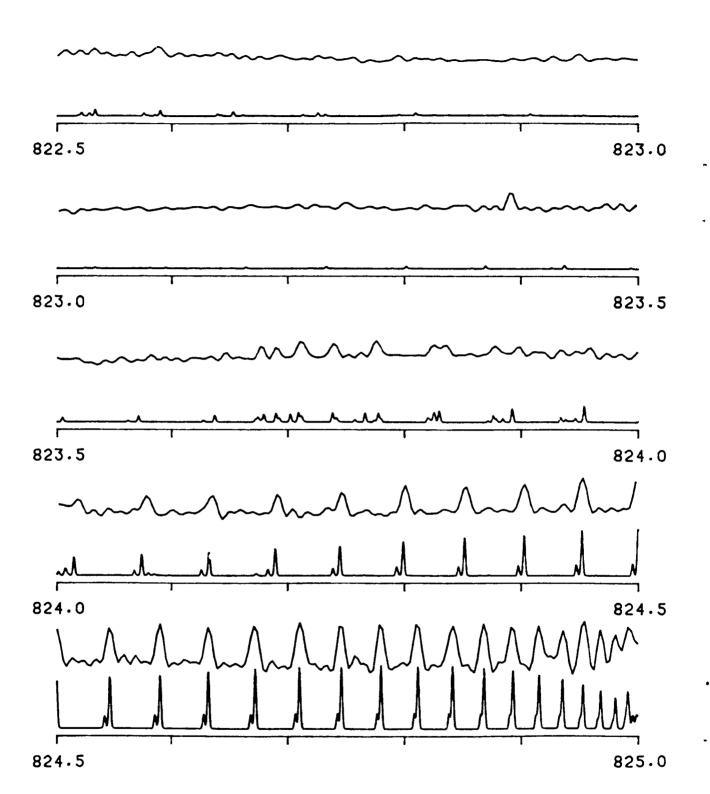


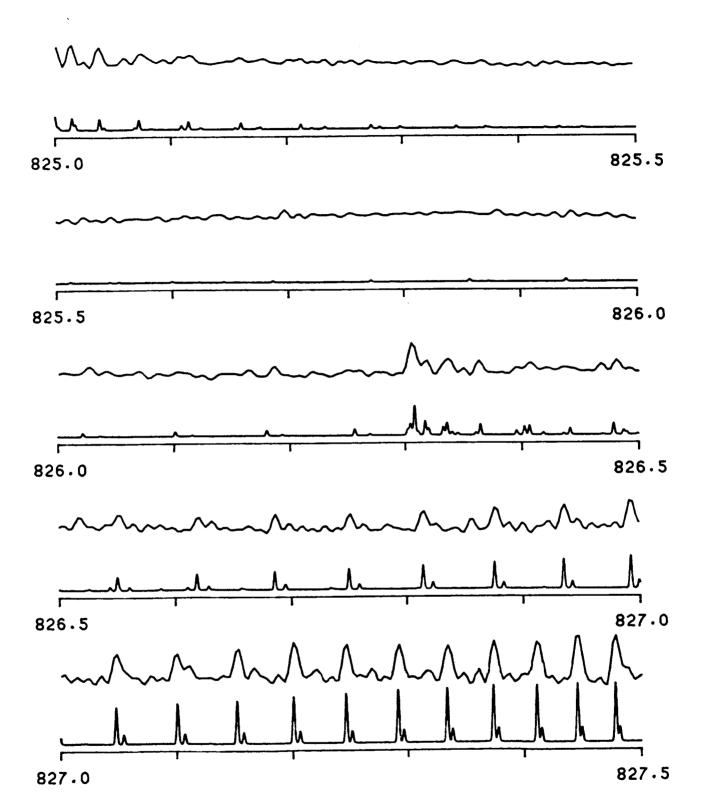


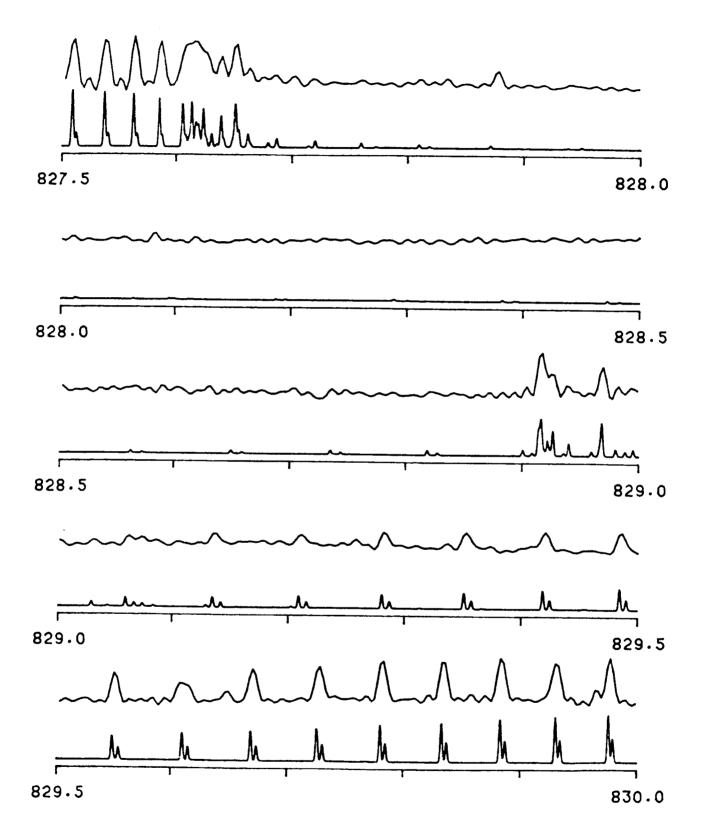


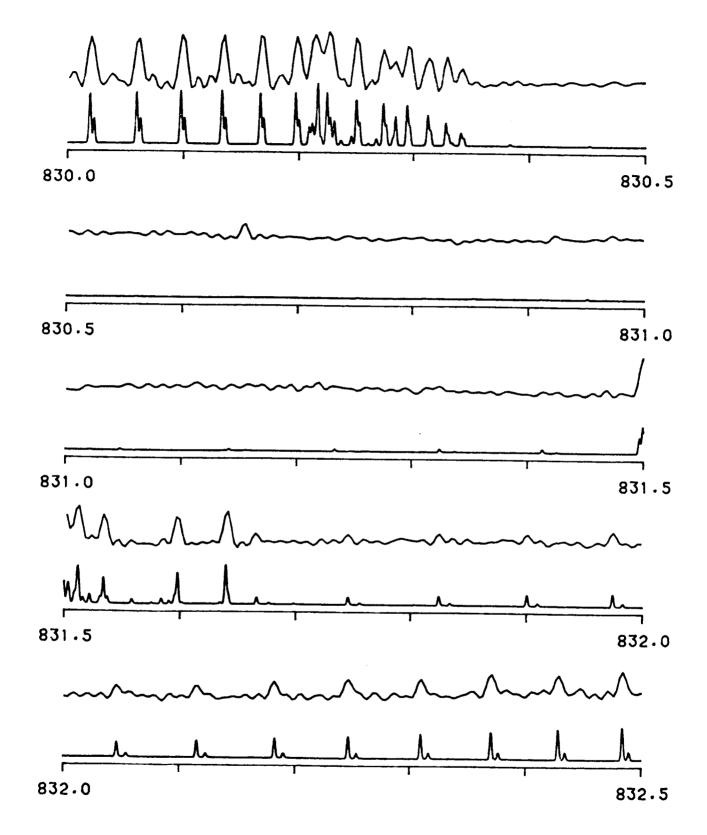


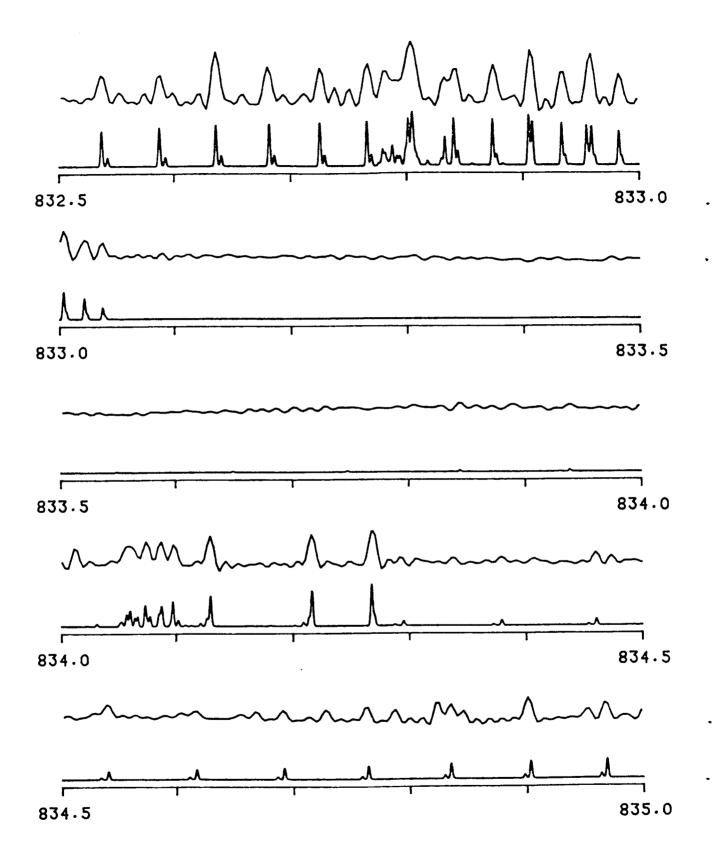


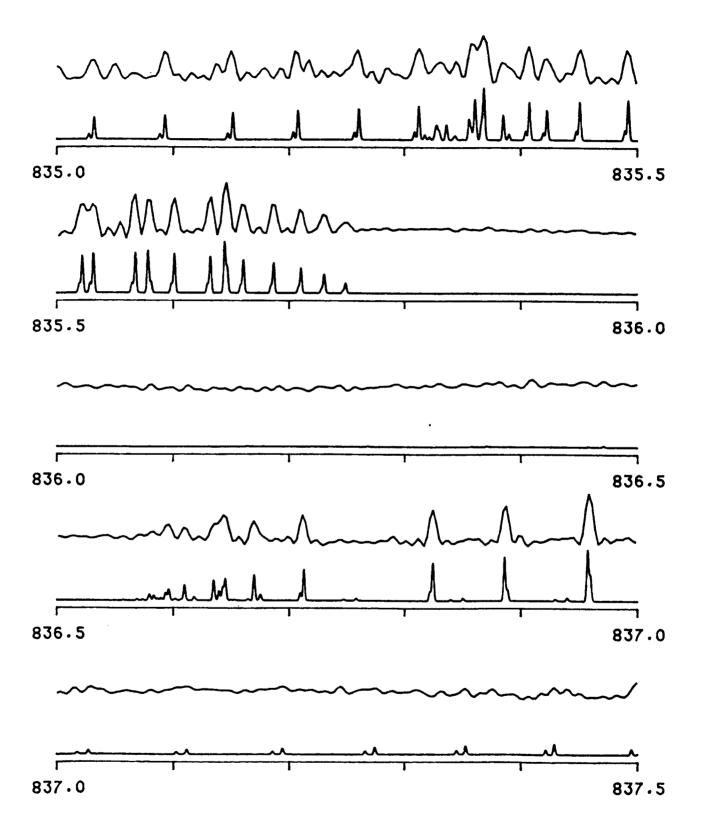


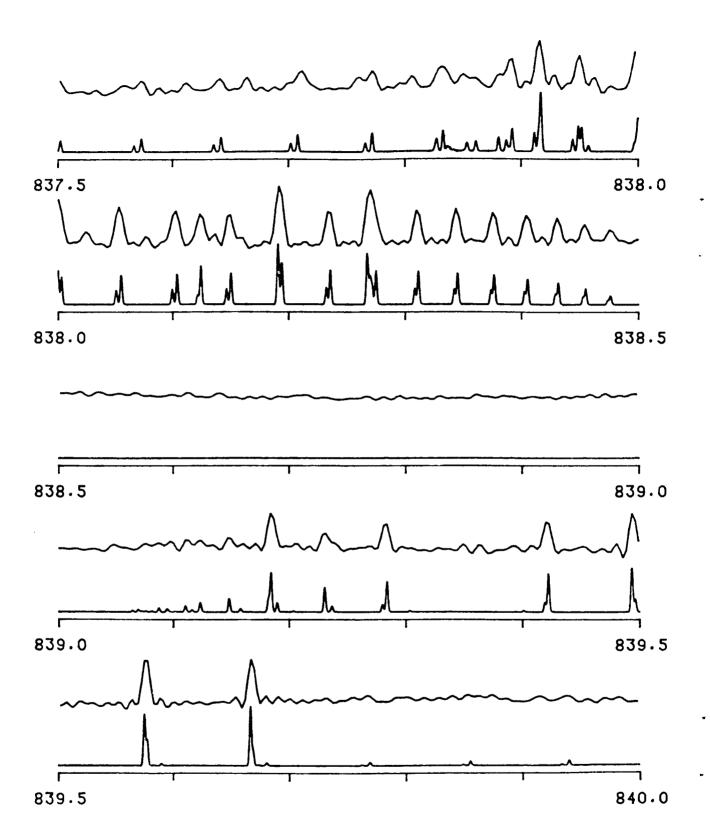


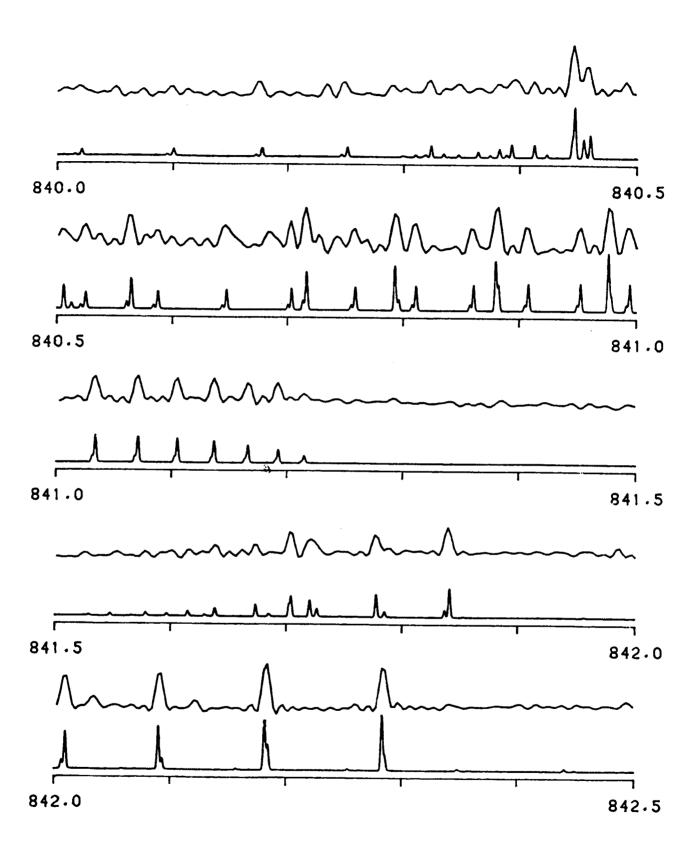


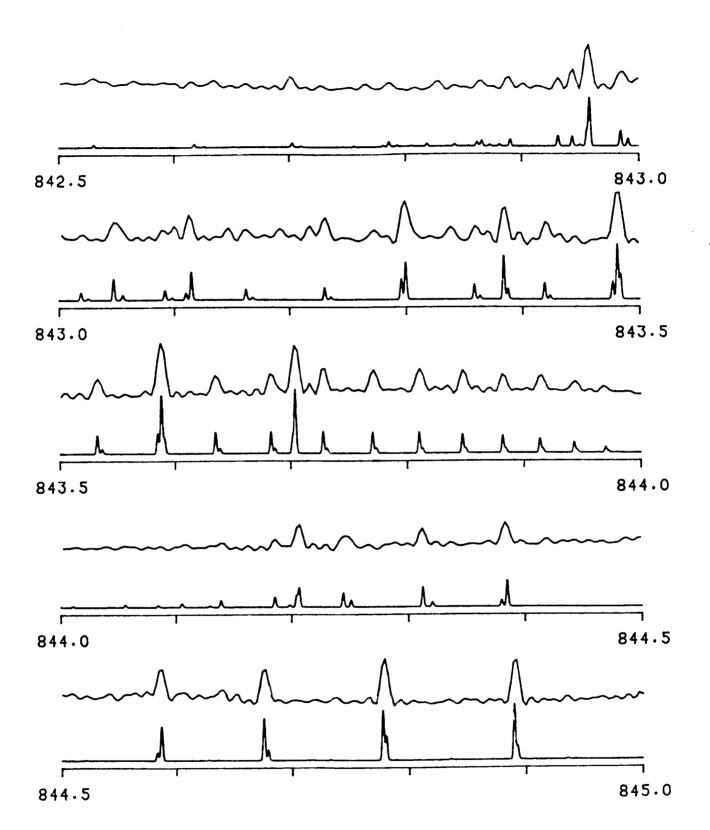


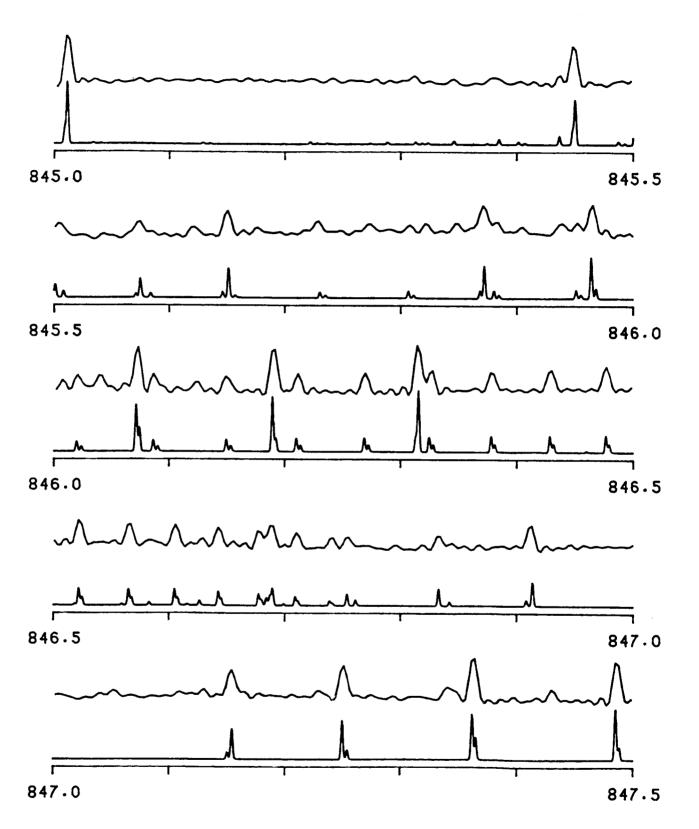


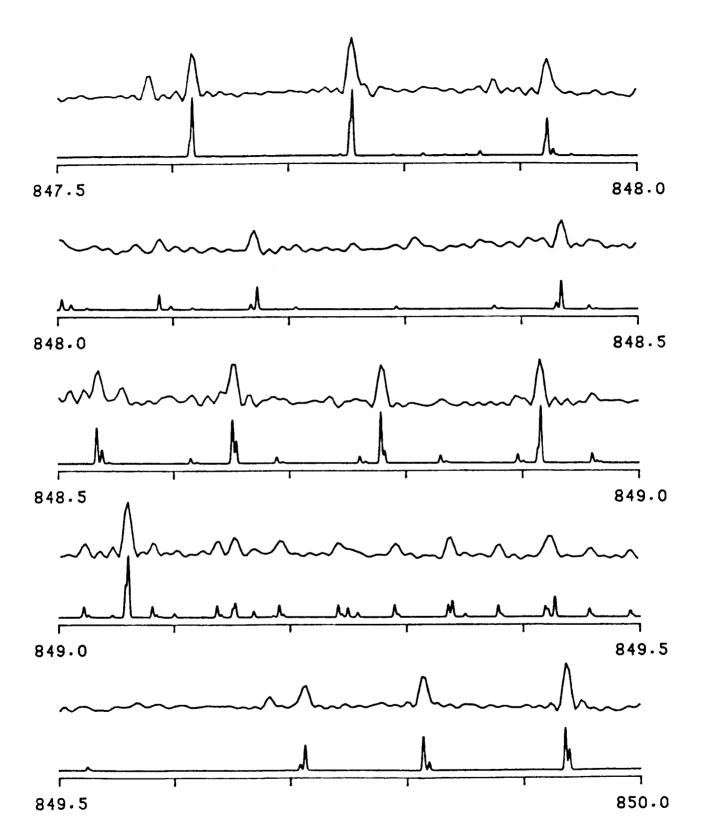


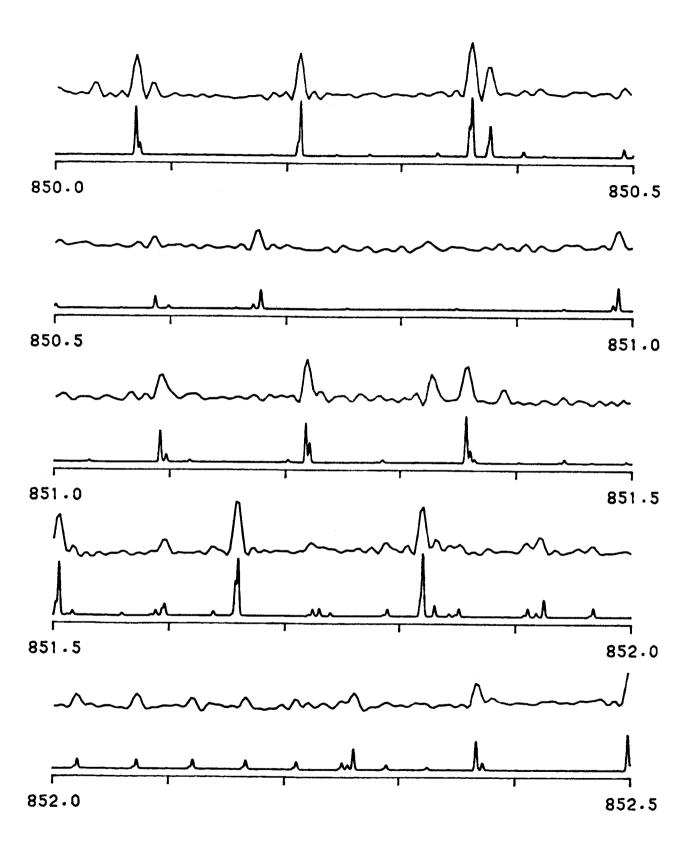


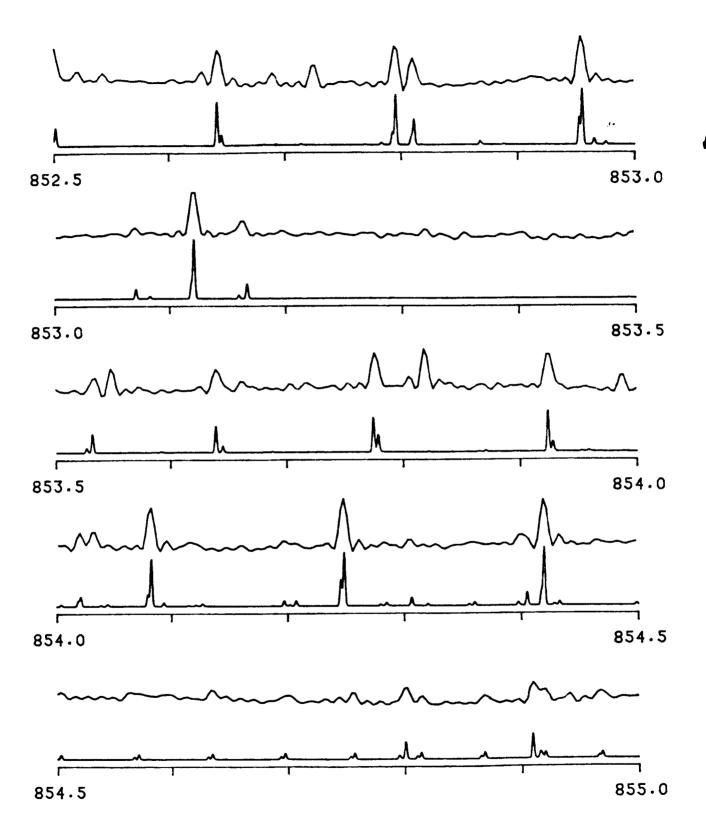


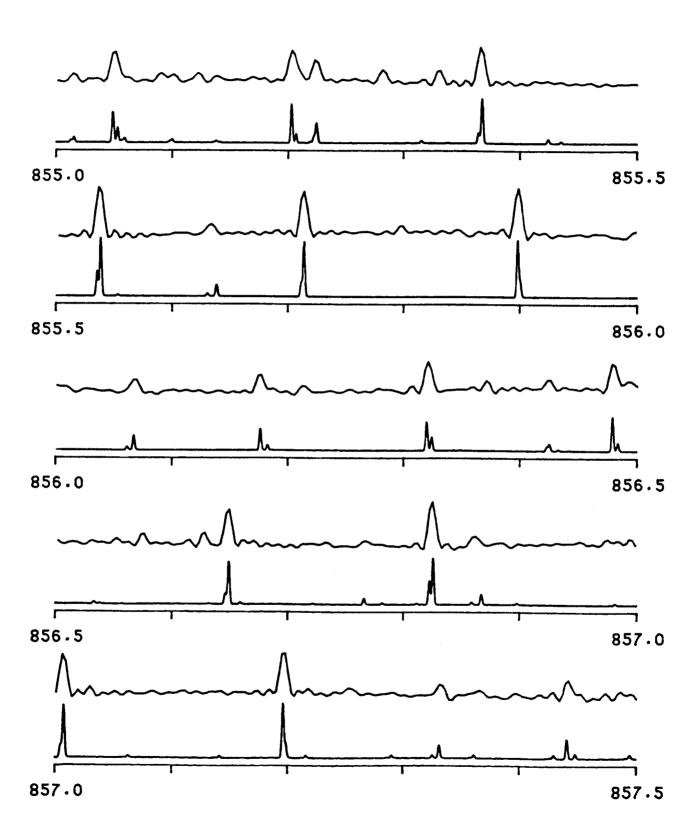


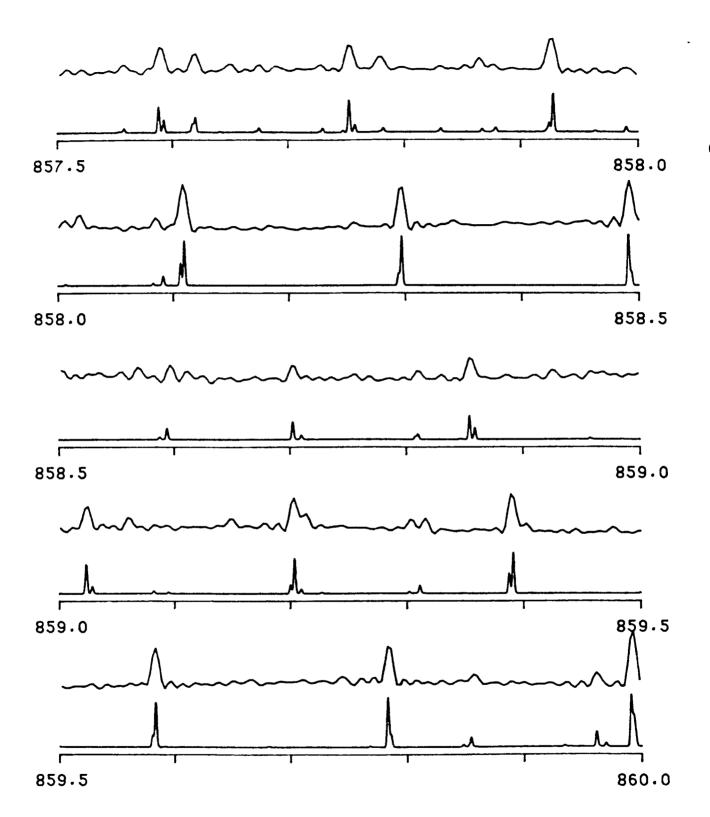


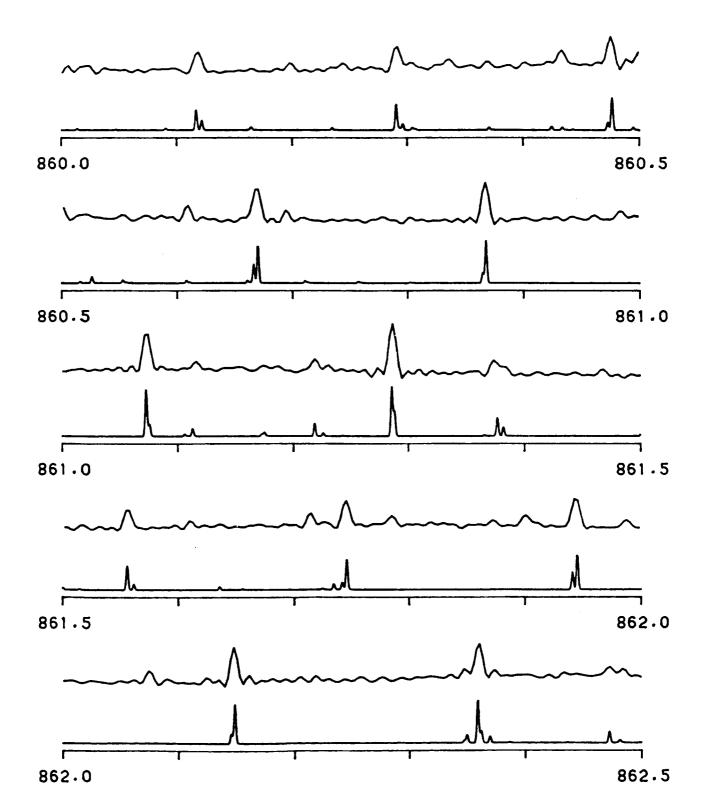


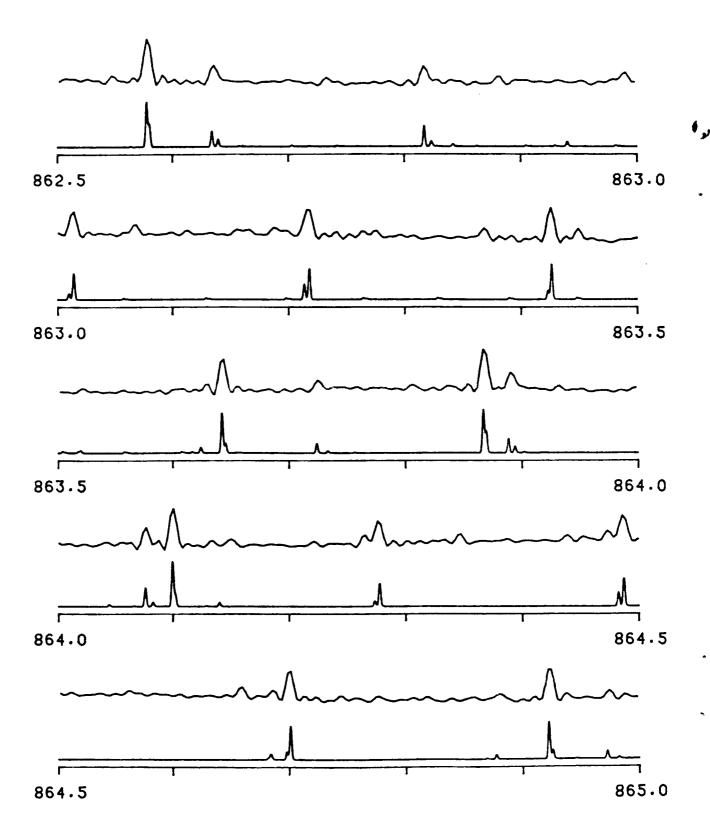


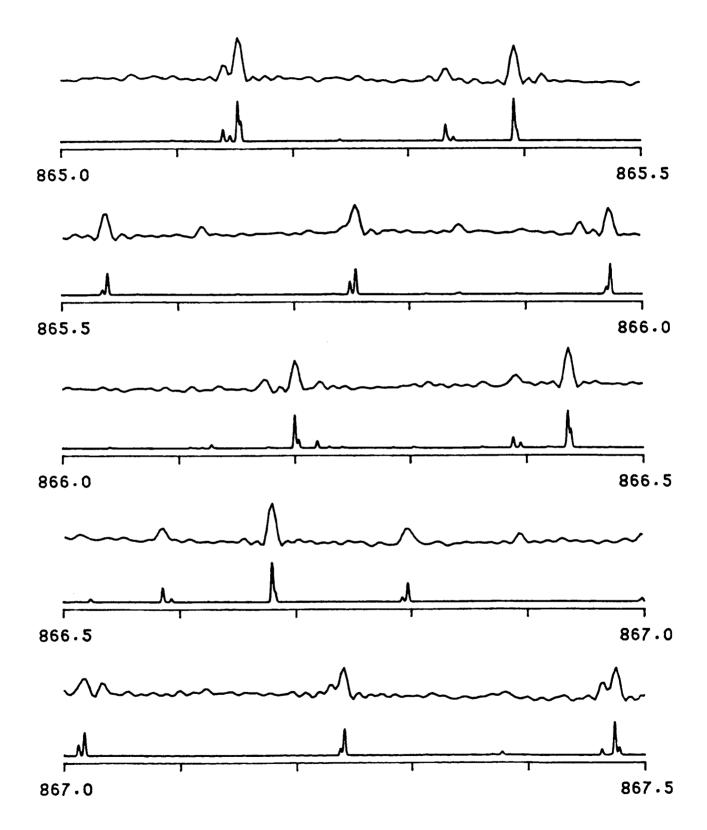


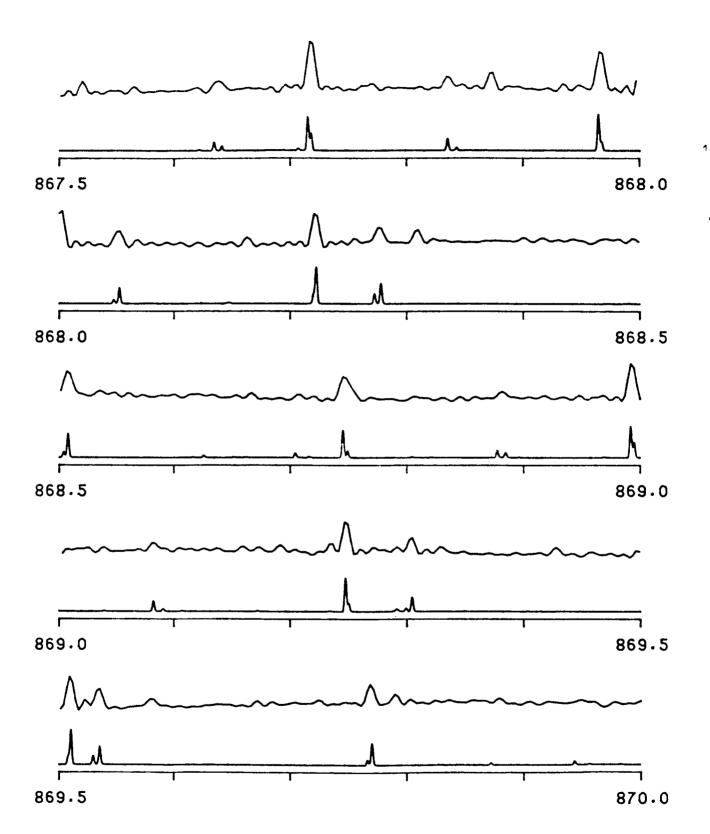


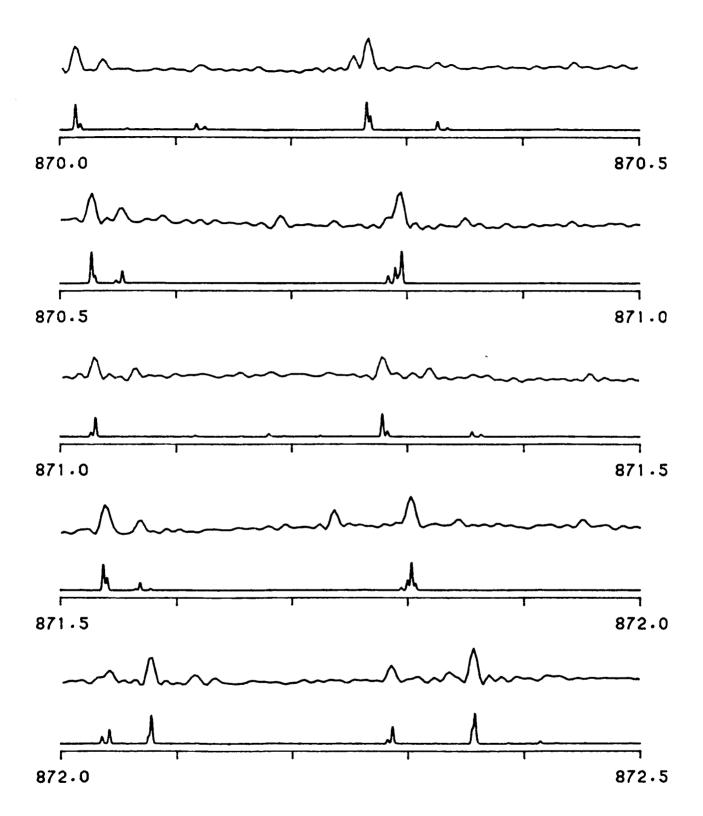


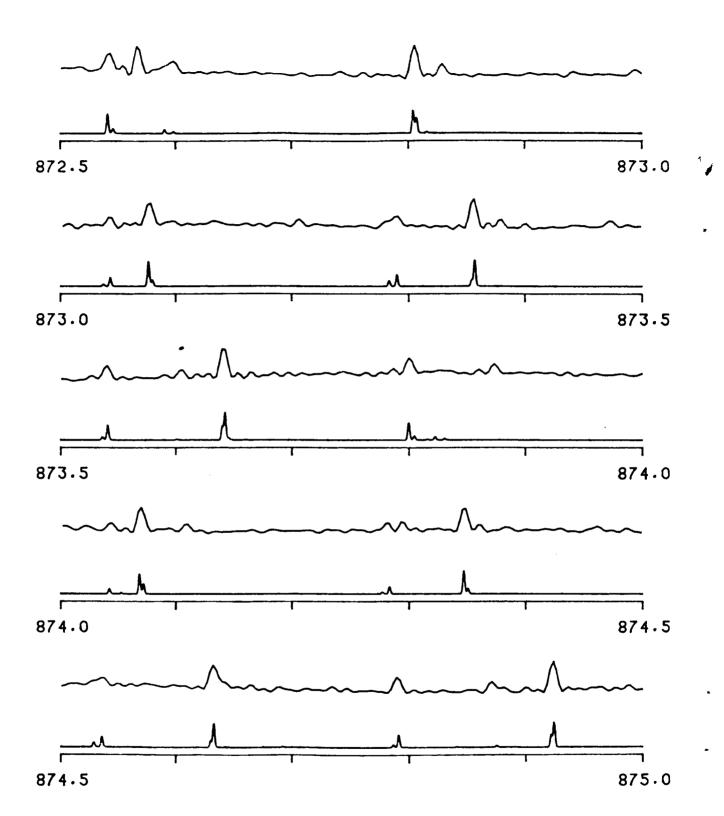


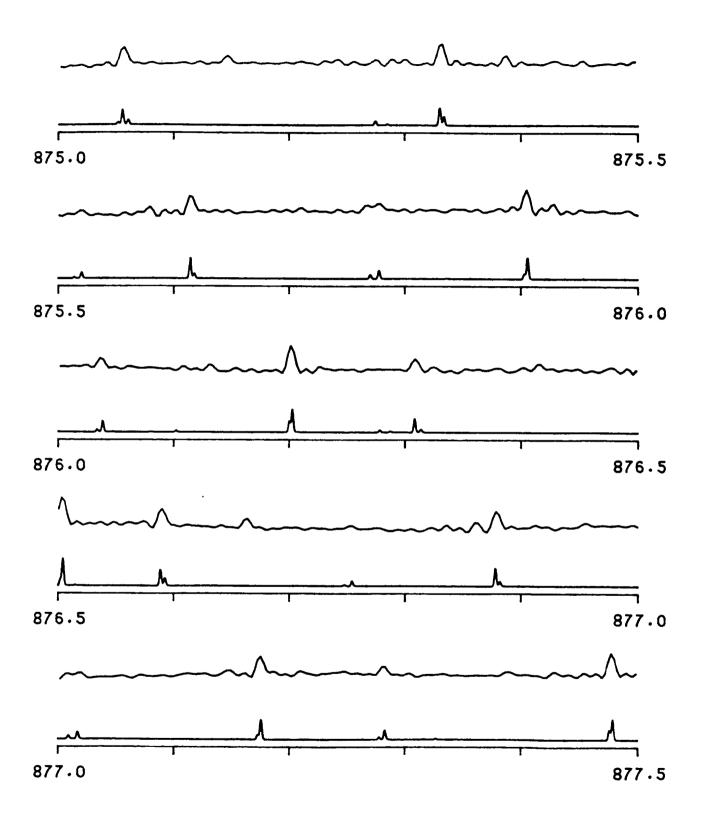


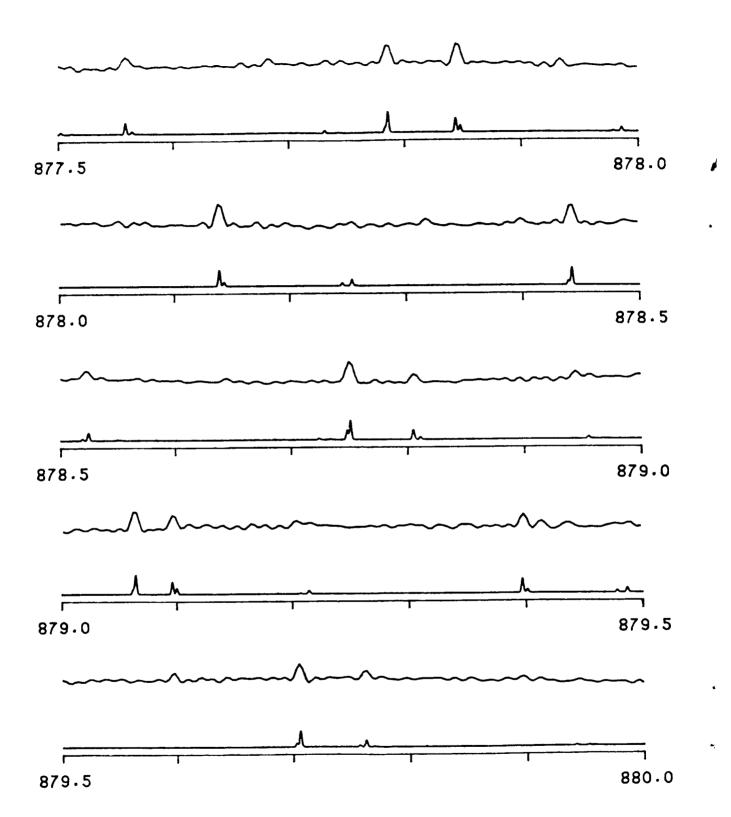


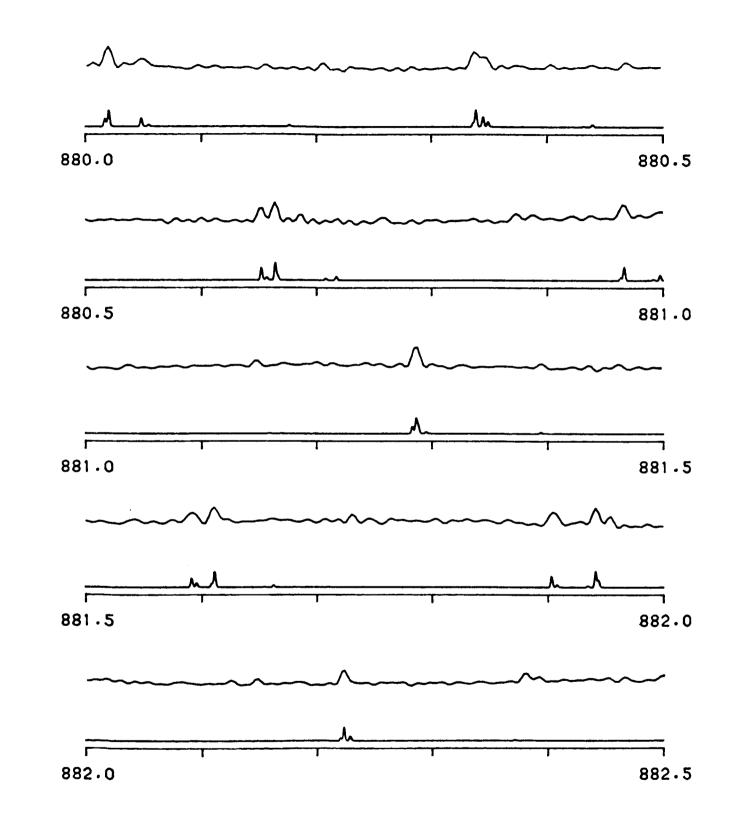




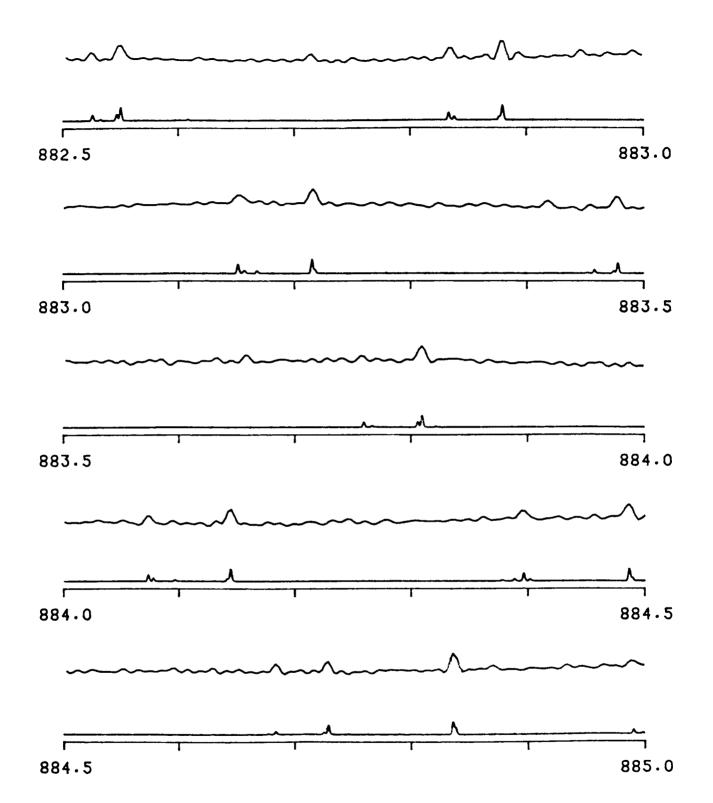


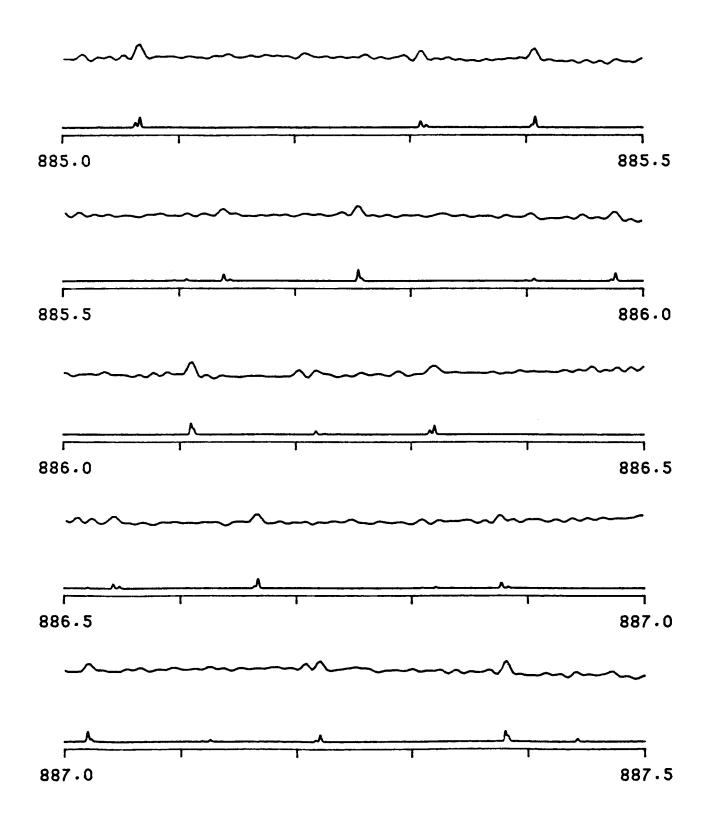


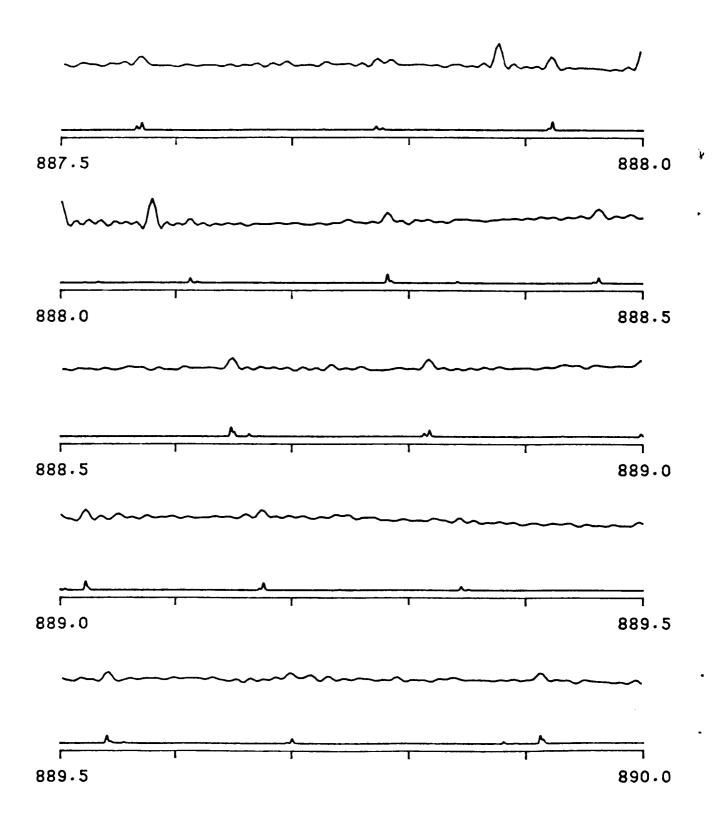


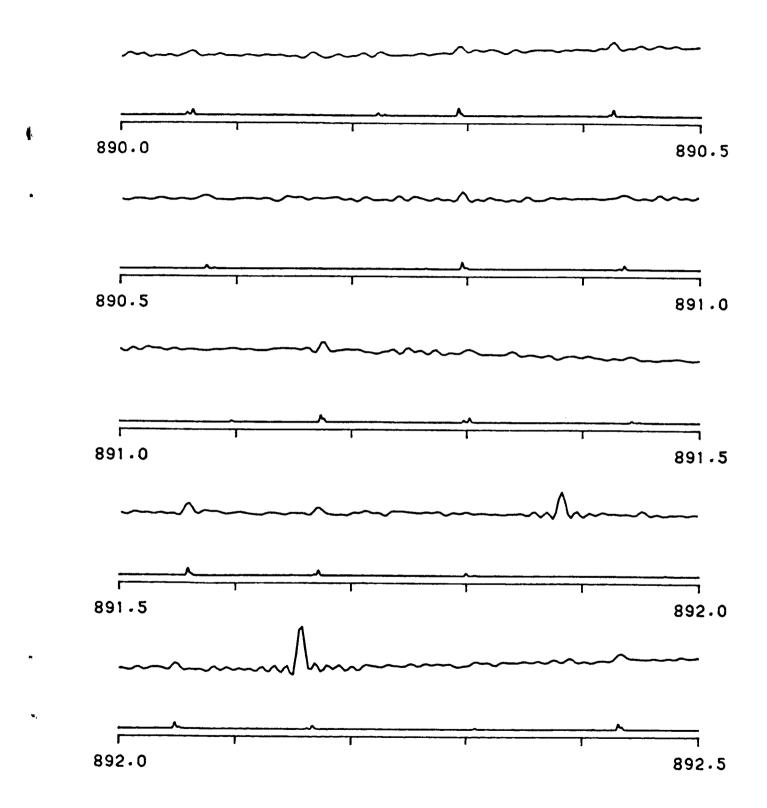


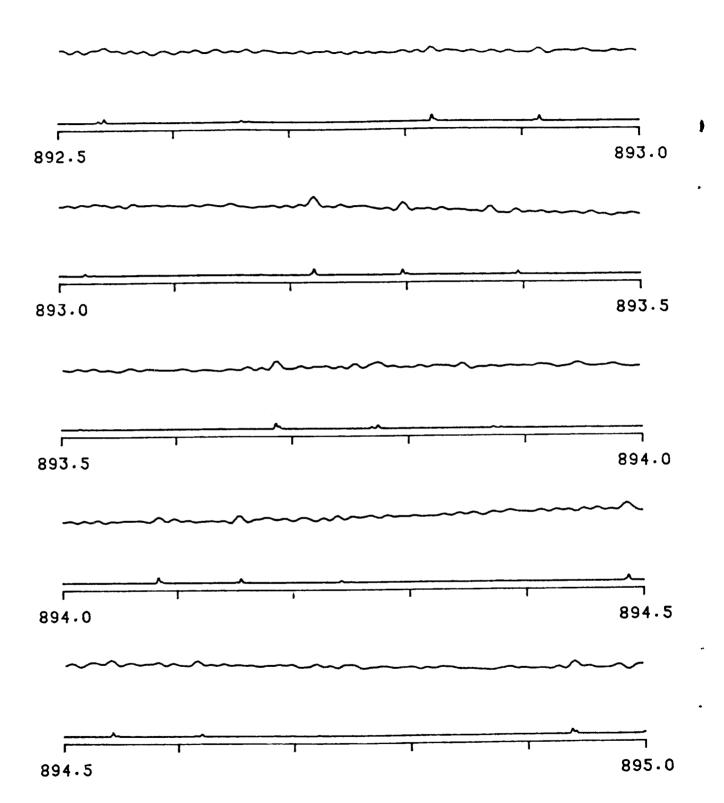
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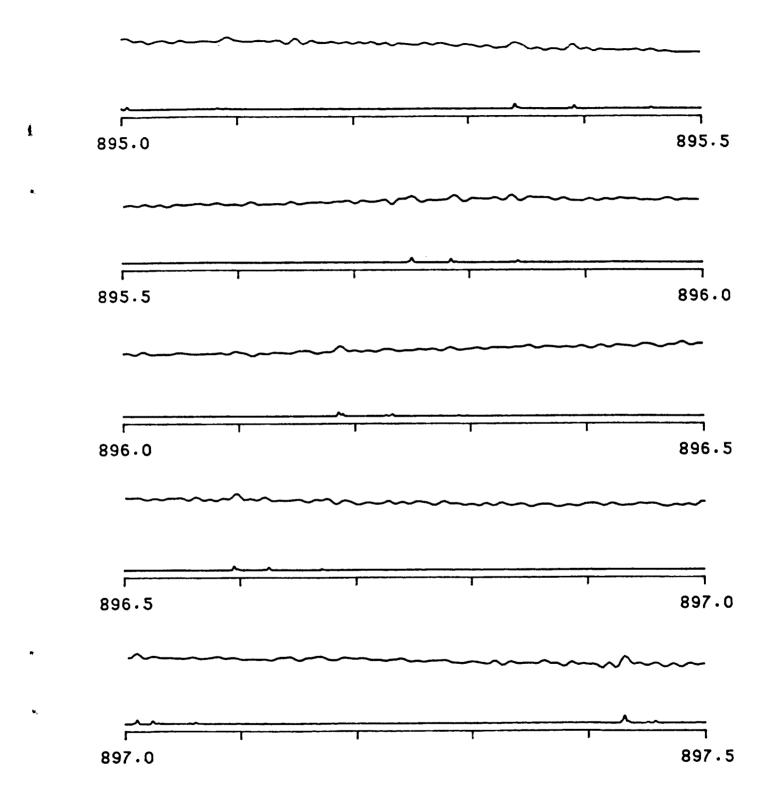












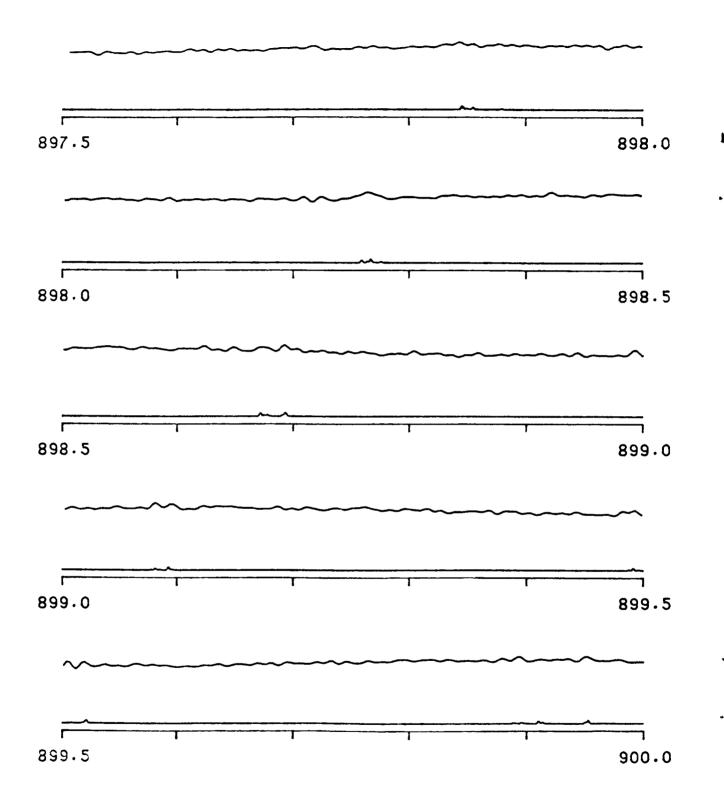


Table 1

Assignment, calculated transition (cm $^{-1}$) and calculated relative intensity from 765 cm $^{-1}$ to 900 cm $^{-1}$ for all transitions such that K < 30 and J < 50. The analysis used transitions $^{-14}$ < K Δ K < $^{+19}$ and J < 41. Extrapolation beyond the experimental transitions are included since some added verifications have been obtained $^{-14}$ < K Δ K and since the calculated transition frequencies are probably better than any other current "guesses".

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.00026 .000104 .52007 .26003 .26003 .18528 .000632 .000334 .00013 .000334 .000334 .000334 .000334 .000334 .00034 .000272 .000273 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 .00060 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	899	899	895	899.							
K J S	7 7	3	0 #	41 1				0 0			
×				7 6							
				RR							
I N I	0.03144	0.00170	0.00680	0.00994	0.03976	0.08992	0.02248	0.01072	0.03198	0.12792	0.00268
WAVE NO	.09631	.13330	.14386	899.48157	.48761	.49153	.49638	.50849	.51786	.52059	.52158
×	899	899	899	899	899	899	899	899	899	899	899
S S	0 77	15 3	15 1	36 3	9	2 6	0 6	3 2	23	1 2	3 0
¥	16 2	7	7	11 3	113	14 2	14 2	7 7	17 2	17 2	8
	R	RR	RR	RR	RR	RR	RR	RR	RR	R R	RR
IN1	0.07107	0.14213	0.02080	898.70178 0.01040	0.00207	0.00413	0.01926	0.07704	0.02944	0.12576	0.00736
0 <u>N</u>	9093	9337	3397	0178	7933	3715	1684	3137	3531	7820	1457
WAVE	898.6	898.69	898.69	398.70	398.7	398.79	899.0	899.00	899.08	399.09	399.00
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→	9 19	3 19	07 6	04 6	5 47	2 47	3 31	3 31	38	5 24	38
_				RR							
LN.	0.00003	0.00007	0.00003	1 0.00007	0.00003	0.03157	0.11867	0.05933	0.06313	0.00007	0.00003
WAVE NO	8.39198	8.48854	8.49115	898.58511	8.58764	8.67140	8.67201	8.67544	8.67795	8.67889	8.68134
	1 89	3 89	1 89	3 89	1 89	2 89	3 89	1 89	98	3 89	1 89
S L	33	32	32	31	31	33	. 92	. 92	33 (30	30
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This document is concerned with the high resolution laboratory absorption spectrum of the 12µm band of ethane gas. The data were obtained using the McMatl Solar Telescope 1-metre Fourier Transform interferometer at Kitt Peak National Observatory and tunable diode laser spectrometers at the University of Tennessee and NASA/Goddard Space Flight Center. Over 2000 individual vibration-rotation transitions were analyzed taking into account many higher order effects including torsional splitting. Line positions were reproduced to better than 0.001 cm ⁻¹ . Both ground and upper state molecular constants were determined in the analysis. Part I of this document contains a discussion of the experimental details, the analysis procedures and the results. A list of ethane transitions occurring near ¹⁴ CO ₂ laser lines needed for heterodyne searches for C ₂ H ₆ in extraterrestrices.									
Part II contains a spectral catalogue of the ethane vo fundamental from									
$765~\rm cm^{-1}$ to 900 cm $^{-1}$. The contents include: (a) a high dispersion (1 cm $^{-1}$ / 12 in.) plot of both the Kitt Peak interferometric data and a simulated spectrum with Doppler-limited resolution using the model of Part I; (b) a table of over 8500 calculated transitions listed quantum number assignments, frequencies and intensities.									
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